
Direct imaging elucidates ionic memory in two-dimensional nanochannels

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Supplementary Information

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Supplementary Note 1. Fabrication of 2D nanochannels

The fabrication of angstrom-channel devices (Supplementary Fig. 1a) involves stacking individual layers of 2D materials, such as graphene, hexagonal boron nitride into a stack on a 500 nm thick double-sided polished silicon nitride on silicon (SiN_x) substrate. We followed a modified version of the fabrication process previously reported^{1,2} and is outlined below (Supplementary Fig. 1b).

The nanochannel device in this study features a trilayer stack, where spacers are sandwiched between bottom and top flakes made of 2D materials. A bottom hexagonal boron nitride (hBN) flake is transferred onto a silicon nitride (SiN_x) free-standing membrane with a micro-hole of $\sim 3 \times 25 \mu\text{m}$. A few layer graphite flake was then patterned using electron beam lithography, followed by dry etching to be used as a spacer layer consisting of an array of parallel line-shaped strips that serve as nanochannels for ion transport. Supplementary Fig. 1c shows atomic force microscopy (AFM) image of a typical spacer. The height of the spacer from the AFM image is $h \sim 1.7 \text{ nm}$ which corresponds to 5-layer graphene and the channel width is $\sim 100\text{-}110 \text{ nm}$ and each of the channels is separated by $\sim 1 \mu\text{m}$. To confine the fluid, another top hBN flake was transferred onto the spacer flake, which was then transferred together onto the back-etched bottom flake on SiN_x membrane to achieve a top-spacer-bottom trilayer stack. To define the channel length, a rectangular aperture was patterned on to the trilayer stack by photolithography at a specific distance ($L \approx 5 \mu\text{m}$), followed by dry etching. The spacers, arranged as an array of parallel strips, serve as channels for ion transport. We chose hBN as the top material for its transparency, which allows light to pass through, providing insights into the dynamics within the trilayer stack.

Supplementary Note 2. Imaging setup

In-operando thin film interferometry measurements were performed using the experimental setup illustrated in Supplementary Fig. 2. Imaging was carried out using a water-dipping objective integrated into a custom-designed fluidic cell. Given that the dimensions of the nanofluidic chips were smaller than the water-dipping objective, a PEEK adapter (square with a 1 mm aperture) was fabricated to support the chip, which was affixed using epoxy resin (Araldite Standard). The chip was mounted in an upward orientation to facilitate imaging the trilayer stack on the SiN_x substrate.

This adapter was positioned between the two components of the PEEK fluidic cell, which were secured by threaded connections and sealed using O-rings. The bottom chamber of the cell included a leak-proof opening to accommodate the bottom electrode, whereas the top chamber remained open to allow immersion of both the objective and the top electrode.

A Nikon CFI Plan 100X ($NA=1.1$) water-dipping objective with a 2.5 mm working distance was employed for imaging. The objective-to-sample distance was initially adjusted using a coarse z-positioner, followed by manual sample positioning via an xy translation stage. Fine adjustments for optimal focus were achieved using a 3-axis piezoelectric stage.

The sample was illuminated using a thermal white light source with a 10 nm bandpass filter having a central wavelength of 635 nm and was attenuated using neutral density (ND) filters to achieve appropriate illumination power densities. The collimated beam was then focused onto the back focal plane of the objective using an achromatic doublet lens.

The objective was mounted at the base of a filter cube mount containing a 50/50 non-polarizing beam splitter. This configuration allowed 50% of the illumination light to be directed toward the sample, while the remaining 50% of the reflected light was transmitted towards the camera. Image acquisition was performed using a Hamamatsu ORCA Flash 4.0 CMOS camera, with an achromatic doublet lens employed for image formation.

Supplementary Note 3. Electrokinetic Measurements

Electrokinetic measurements were performed using Ag/AgCl electrodes to apply the potential and record the resulting current. The setup included a FEMTO amplifier (DLPCA-200) and a digital/analog converter (NI 63 series), which also synchronises the electrokinetic data with optical imaging. For datasets used in Figs. 3-5 and Supplementary Figs. 21 and 23, electrokinetic measurements were temporally binned to match the acquisition rate of the optical imaging data. This resulted in a temporal sampling rate ranging from 0.5 to 10 frames per second (fps), depending on the acquisition rate of the corresponding optical imaging dataset. The downsampled electrokinetic data were used to generate the $I-V$ curves in Fig. 3 and the conductance-versus-time traces presented in Figs. 3–5 and Supplementary Figs. 21 and 23. Also, conductance traces presented in Figures 3, 4, 5 and Supplementary Figure 21, 23 were obtained by dividing current by voltage, omitting points near zero voltage with ill-defined conductance. The conductances $G_{\min}$ and $G_{\max}$ used to define the normalized conductance δG

were computed as the 0.1 and 0.9 quantiles of the conductance trace to average out the cycle-wise variability of the conductance minima and maxima.

Supplementary Note 4. Image acquisition and processing

In the case of drift during the acquisition, the image was stabilized using the image stabilizer plugin for ImageJ by Li and Kang³. The image was initially acquired with a 2×2 binning, followed by an additional 2×2 binning applied during data analysis.

Supplementary Note 5. Transfer matrix calculation of the contrast

For small deformations, the VIC Ψ corresponding to a blister of height $h_B(\Delta V)$ reads:

$$\Psi(\Delta V) = 4\pi n_w h_B(\Delta V) / \lambda \cdot F(n_j, t_j, \lambda) \quad (0)$$

where n_w is the refractive index of water and $F(n_j, t_j, \lambda)$ is a dimensionless parameter of order 1 which depends on the layer thicknesses t_j and refractive indices n_j of the layers $j = 0, \dots, 5$. $F(n_j, t_j, \lambda)$ defines the deformation sensitivity of the experiment, and we chose $\lambda = 635$ nm to maximize it for the relevant device parameters. We detail below the full transfer matrix method (TMM) contrast calculation for the 6-layer thin film heterostructure. For all experimental geometries, we quantified the impact of layer thickness uncertainty derived from the ~ 10 nm window of our exfoliation metrology, which results in an uncertainty $\Delta h_B/h_B = \Delta F/F \sim 30\%$ for contrast-based height estimates. This bound ensures that the contrast remains monotonic for small deformation magnitudes across all devices (Supplementary Figs. 3-5).

5.1 Simulation details

The 6-layer thin film reflectance is evaluated using the TMM. We consider normal-incidence reflection from a multilayer structure immersed in water, with the following stack (from top to bottom):

1. Semi-infinite water: $n_0 = n_w$
2. Top hBN: thickness t_{top} , refractive index $n_1 = n_{hBN}$
3. Water blister: thickness h_B , refractive index $n_2 = n_w$
4. Bottom hBN: thickness t_{bot} , refractive index $n_3 = n_{hBN}$
5. Silicon nitride (SiN): thickness t_{SiN} , refractive index $n_4 = n_{SiN}$
6. Semi-infinite water: $n_5 = n_w$

Our goal is to compute the reflected intensity $\eta(h_B(\Delta V))$ to obtain the VIC. We assume no losses in the media and that the resulting voltage-induced intensity modulations arise purely from

interference. Moreover, we model the case of blisters much wider than the light wavelength, neglecting refraction effects at the edges.

Interface and propagation matrices

Interface and propagation through the layers of the heterostructure can be formalized through transfer matrices acting on the electric field vector, $\begin{pmatrix} E^\downarrow \\ E^\uparrow \end{pmatrix}$ where $E^\downarrow$ corresponds to the downward-propagating wave and $E^\uparrow$ to the upward-propagating wave.

At each interface between refractive indices n_i and n_j , we define the reflection coefficients $r_{ij} = (n_i - n_j) / (n_i + n_j)$ and transmission coefficients: $t_{ij} = 2n_i / (n_i + n_j)$.

An interface from medium i to j contributes an interface matrix I_{ij} , and a layer of thickness t_L and index n contributes a propagation matrix $P(n, t_L, \lambda)$, respectively given by:

$$I_{ij} = \frac{1}{t_{ij}} \begin{pmatrix} 1 & r_{ij} \\ r_{ij} & 1 \end{pmatrix} ; \quad P(n, t_L, \lambda) = \begin{pmatrix} \exp(2i\pi n t_L / \lambda) & 0 \\ 0 & \exp(-2i\pi n t_L / \lambda) \end{pmatrix}$$

We now construct the full transfer matrix $M(h_B)$ for the 6-layer system. The light is incident from the top (semi-infinite water), and propagates through each interface and layer in sequence.

$$M(h_B) = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} = I_{01} \cdot P(n_1, t_{top}) \cdot I_{12} \cdot \mathbf{P}(\mathbf{n}_2, \mathbf{h}_B, \lambda) \cdot I_{23} \cdot P(n_3, t_{bot}) \cdot I_{34} \cdot P(n_4, t_{SiN}) \cdot I_{45}$$

In this expression, we highlighted $\mathbf{P}(\mathbf{n}_2, \mathbf{h}_B, \lambda)$ which is the only blister-dependent part.

Reflected Intensity $\eta(h_B)$

In the absence of incoming light from below (boundary condition $E^\uparrow = 0$ in the bottom water), the reflection coefficient $r(h_B)$ is given by: $r(h_B) = M_{21}(h_B) / M_{11}(h_B)$

This yields the reflected intensity as: $\eta(h_B) = |r(h_B)|^2$ and thus the contrast through equation (1).

5.2 Simulation results

Results are shown in Supplementary Figure 3, with the aim of demonstrating that semi-quantitative topographical information can be retrieved from contrast maps *in the device geometries used in this work*. The two main geometrical parameters that can be set experimentally are the bottom and top thicknesses t_{bot} and t_{top} chosen during the van der Waals heterostructure fabrication. We examined the parameter range in which the contrast scales monotonously with the blister height h_B .

Supplementary Figure 3d presents the TMM-evaluated contrast for a range of blister heights from 1 nm to 80 nm. We find that larger blisters provide an increasing contrast, which can be

either negative (blue) or positive (red), depending on the region of the parameter space $\{t_{bot}, t_{top}\}$ chosen during fabrication. Some parameter choices result in reflectance cancellation, as shown in Supplementary Fig. 3b. These regions should be avoided, as the contrast then provides ambiguous results over the device topography. For this reason, the light wavelength and device geometry should be chosen to avoid this pathological case. Thus, we used $\lambda=635$ nm and we operated in the geometry range given by the yellow square in Supplementary Figure 3d ($t_{bot}=50\pm5$ nm and $t_{top}=90\pm10$ nm), for which the blister formation always results in a positive contrast that scales monotonously with the blister height (Supplementary Fig. 3b). The contrast-to-height relationship is sub-linear but can be approximated by a linear increase near zero, which defines the deformation sensitivity F , as defined in equation (0). The distribution of F values in the used geometry range has a median value of 1.55 with extremal values of 1.40 and 2.27 (Supplementary Fig. 3c). This shows that for small deformations, the VIC gives semi-quantitative information over the blister height within a factor ~ 2 and can be made fully quantitative by accurate AFM height measurements of the device.

Supplementary Note 6. Device Endurance

All devices listed in Supplementary Table 1 were measured multiple times, with each measurement having at least 2 cycles. We performed voltage amplitude sweeps on device 1 in the range of 50 mV to 1 V at 10 mHz and 100 mHz, followed by frequency variation studies. For device 2, we were able to complete the full experimental set, including amplitude, frequency, concentration, and salt-dependence studies and repeated the entire measurement three times to ensure reproducibility. All other devices were treated with at least 1 M KCl multiple times.

Supplementary Table 1

Device Number	Concentration	Species	Configuration			Height (nm)	Annealed (Yes/No)	Comments
			Top	Spacer	Bottom			
Device 1	1 M	KCl	hBN	Gr	hBN	1.70	No	
Device 2	1 mM - 1 M (KCl)	KCl, NaCl, LiCl, CaCl ₂	hBN	Gr	hBN	0.68	No	
Device 3	1 mM - 1 M (KCl)	KCl, NaCl, LiCl, CaCl ₂ , AlCl ₃	hBN	Gr	hBN	1.02	No	
Device 4	1 mM - 1 M	KCl, CaCl ₂ , AlCl ₃	hBN	Gr	hBN	0.68	Yes	
Device 5	1 M	KCl	hBN	Gr	hBN	1.70	Yes	Gold patch
Device 6	1 M	KCl	hBN	Gr	hBN	1.02	Yes	Gold patch
Device 7	1 M	KCl	hBN	hBN	hBN	50	Yes	Thick channel
Device 8	1 M	KCl	hBN	Gr	hBN	1.02	Yes	
Device 9	1 M	KCl	hBN	Gr	Gr	1.7	No	Bottom Gr
Device 10	1 M	KCl	hBN	Gr	Gr	0.68	No	Bottom Gr
Device 11	1 M	KCl	hBN	---	hBN	0	Yes	
Device 12	1 M	KCl	---	---	hBN	---	No	hBN on SiNx
Device 13	1 M	KCl	---	---	hBN	---	No	hBN on SiNx
Device 14	1 M	KCl	hBN	Gr	Gr	1.02	No	Bottom Gr

Supplementary Note 7. Theoretical modelling of the observed deformations

7.1. Blister modelling and energy barrier

7.1.1 Energy functional

We consider a nanochannel of hBN of length $L = 5 \mu\text{m}$ and thickness $t_L \sim 100 \text{ nm}$ with strong confinement of $h = 5 \text{ nm}$ in which we use an electrolyte at ionic concentration of $\rho_s = 1 \text{ M}$ and apply on it a voltage drop of $\Delta V = 1 \text{ V}$. Typical blisters have a height of $h_B \sim 10 \text{ nm}$ over a radius $R \sim 1 \mu\text{m}$.

The blister is an elastic deformation modelled by a deflection of height h_B , whose energy cost is controlled by the Young's modulus of hBN out-of-plane $Y_{\perp} \approx 30 \text{ GPa}$ and the in-plane $Y_{\parallel} \approx 800 \text{ GPa}$. In practice, the effective Young's modulus intervening in the bending energy of multi-layer hBN decreases with the number of layers: $Y_{\perp}^{\text{eff}} \approx Y_{\perp}/N$ where N is the number of layers, so that the bending is negligible for the experimental configuration⁴. Moreover, the hBN layer lays on spacers in graphene, with which it has an adhesion energy. A blister which covers several nanochannels needs to break its adhesion over the area of contact with the spacers. However, this area can be reduced by already existing bubbles (introduced for instance, during fabrication) which do not break the tightness of the van der Waals assembly. We denote Γ as the adhesion energy per unit surface of hBN on graphene⁵. Overall, the total energy of a hemispheric blister of radius R and height h_B experiencing a pressure P is then given by:

$$\Delta E \approx \alpha \pi R^2 \Gamma + (\pi Y_{\perp} t_L / 12 R^2) h_B^4 - (P \pi R^2 / 2) h_B \quad (1)$$

The first term is the adhesion energy to break. The second term is the stretching energy of the deformed hBN layer. The last term is the work of the applied pressure.

7.1.2 Blister energy threshold

For the blister to form, the first step is to overpass the energy barrier of adhesion. In practice, both h_B and R grow together. However, $h_B(t)$ is a growing function of time which is limited by the stretch energy cost. Minimising the energy over h_B leads to this limit deflation:

$$h_B^0 = [3PR^4 / (2Y_{\perp} t_L)]^{1/3} \quad (2)$$

As long as $h_B < h_B^0$, the height grows and the energy cost of expanding the blister (increasing R) is decreasing. Hence, the blister will start expanding at some point unless it remains energetically unfavourable when the height h_B has reached its limit value h_B^0 . It is therefore sufficient to focus on the energetics in the limit case $h_B = h_B^0$.

Thus, the effective energy of the blister reads⁶:

$$\Delta E \approx \pi R^2 [\Gamma - (3/8)(3R^4 / 2Y_{\perp} t_L)^{1/3} P^{4/3}] \quad (3)$$

We immediately notice that $R = 0$, corresponding to the absence of blisters, is a (local) minimum of the energy function. Small blisters are energetically forbidden while large blisters become energetically favourable. In practice, the size of the blisters is however limited by the size of the system. The largest possible blisters, which are the most favorable energetically, then have a size $R \sim L \approx 5 \mu\text{m}$. This explains why the experimentally observed blisters of a size comparable to the system while small blisters are not observed. For these blisters to appear their energy must be negative, which provides a threshold on the required pressure:

$$P_c \approx [(2Y_{ltL})^{1/4} (8\Gamma)^{3/4}] / (3R) \quad (4)$$

Note that this threshold depends on space if the adhesion is not uniform. Even for a single blister, the adhesion might be non-uniform leading to a distribution of thresholds rather than a single value, which would result in the experimental data to a smoothened threshold.

7.1.3 Nucleation

We can also study the energy differential when the blister grows:

$$d\Delta E/dR \approx 2\pi R [\Gamma - (7/8)(3R^4 / 2Y_{ltL})^{1/3} P^{4/3}] \quad (5)$$

which is negative for R large enough. This means that a blister can form only if there is a zone of low adhesion which is large enough. The differential becomes negative for pressure larger than:

$$P_c \approx [(2/3 Y_{ltL})^{1/4} (8/7 \Gamma)^{3/4}] / R \quad (6)$$

which is very similar to Eq. (4).

7.1.4 Hysteresis and memory effect

Once the blister is formed, the adhesion energy vanishes and there is no more threshold to cross. Therefore, when the voltage decreases, the blister size decreases smoothly with:

$$h_B = [3PR^4 / (2Y_{ltL})]^{1/3} \quad (7)$$

When the pressure is below the threshold, the blister remains until the height reaches zero and adhesion is restored. Thus, it keeps memory of the history: whether the threshold has already been crossed or not since the last zero voltage.

7.1.5 Dynamical effects

The previous analysis was purely static: we determined what is the state which minimises the energy. In practice, however, the evolution towards a new mechanical minimum might take some time. In all practical cases, the inertia of the deformed layer remains negligible. We identify two main effects which might be dynamically limiting: the de-adhesion process and the filling of the blister when growing once the threshold has been reached. The former is

known to contain many timescales and is hard to evaluate theoretically⁷. The latter can be estimated roughly.

Once the threshold is reached, the energy landscape pushes the blister deflation towards h_B given by Eq. (7) and the blister grows. Nevertheless, for a nanochannel the hydrodynamic resistance R_h is large. Therefore, ensuring a flow rate requires a strong pressure drop. Assuming for simplicity that the blister has a radius R and no curvature, we estimate the required pressure drop to fill the growing blister as $Q \approx \pi R^2 \partial_t h_B \approx \Delta P / R_h$. This pressure drop reduces the pressure P_c which pushes the blister to grow and acts as a dissipation for the blister dynamics. In the overdamped regime, we then have the dynamical equation:

$$\pi R^2 \Delta P = \pi^2 R^4 R_h \partial_t h_B = -(\pi Y_{\parallel} t_L / 3R^2) h_B^3 + (P_c \pi R^2 / 2)$$

The timescale to grow the blister can then be estimated by

$$\tau \sim \frac{3\pi R^6 R_h}{Y_{\parallel} t_L h_B^2} \sim \frac{36\pi R^6 \eta l}{w h^3 Y_{\parallel} t_L h_B^2}$$

where w is the effective width of the channels, l its length and h its height. Here, for simplicity, we take $w \sim l \sim R$ while $h \approx 1$ nm and $h_B \approx 10$ nm, leading to $\tau \approx 10$ s. Note however that this timescale depends crucially on the detailed environment of the blister: local adhesion distribution through the dynamical history of blister radius and height; distance to the nanochannel edges and neighbouring blisters through the hydrodynamic resistance. Consequently, τ is expected to be blister-dependent and our value to be an approximate order of magnitude.

Thus, while an exhaustive investigation of the timescales at play in these memristors is beyond the scope of this paper, we provided a first rough estimate for the frequency cutoff of the memristor around 100 mHz. We expect the memristive effect to decrease at larger frequencies as a blister has not enough time to grow.

7.2. Bidirectional memristors: Polarisation concentration-induced osmotic pressure

7.2.1 Electroneutrality, resistivity and concentration profile

Since $L \gg \lambda_D$ we assume electroneutrality. Indeed, the electroneutrality will be recovered over the scale of the Debye length thanks to the electric field created by the charge distribution. Thus,

$$c_+ - c_- = -2\Sigma/h \quad (8)$$

According to Nernst-Planck's law, the total flux reads:

$$J_{\text{tot}} = J_+ + J_- = -D \partial_x c_{\text{tot}} - (2\mu\Sigma/h) E(x) \quad (9)$$

and must be uniform. Here, $c_{\text{tot}} = c_+ + c_-$ is the total ion concentration including the surface countercharges. The electric current, including the surface contribution, reads

$$I = J^+ - J^- = c_{\text{tot}}(x) E(x)$$

and must also be uniform. Because the electroneutrality may be locally broken, the electric field may depend on space. This local Ohm's law simply states that the local polarisability accommodates the local electric field to the local ionic resistivity $\rho_i \propto 1/c_{\text{tot}}$. Thus,

$$E(x) = -\Delta V / (c_{\text{tot}} \int dx / c_{\text{tot}}) \quad (10)$$

For a large Dukhin number $Du = |\Sigma|/ehc_0$, the channel becomes selective and the total flux J_{tot} becomes small compared to the electric current I . In this limit, there is a strong polarisation concentration effect and an equilibrium concentration profile. This limit is valid for low salt concentration. For smaller Dukhin numbers the selectivity is partial and the total flux is not negligible. In the experiment, the Dukhin number ranges from intermediate to large values. Nevertheless, we will assume a perfect selectivity in the following in order to allow the model to be tractable and to provide qualitative insights on the physics at play, keeping in mind that we overestimate the effect at large salt concentration. With this assumption:

$$D\partial_x c_{\text{tot}} = (2\mu\Sigma/h)(1/c_{\text{tot}}) (\Delta V / \int dx / c_{\text{tot}}) \quad (11)$$

For simplicity, we denote $\xi = e\Delta V/k_B T$ and $R = \int dx / c_{\text{tot}}$, and we adimensionalise c_{tot} by $2c_0$ and x by L . Then,

$$c_{\text{tot}}(x) = [c_{\text{tot}}(0)^2 + (Du \xi / 2R) x]^{1/2} \quad (12)$$

Also,

$$c_{\text{tot}}(1) = [c_{\text{tot}}(0)^2 + Du \xi / 2R]^{1/2} \quad (13)$$

At the edges the concentrations are not bulk yet due to the concentration polarisation effect. Yet, they are balanced by the diffusion flux between the entrance of the channel and the far away reservoir at bulk concentration. Using symmetric reservoirs and because we must have equal fluxes at each edges, we then obtain:

$$c_{\text{tot}}(0) + c_{\text{tot}}(1) = 2 + 2Du \quad (14)$$

Thus,

$$c_{\text{tot}}(0)^2 + Du \xi / 2R = [2 + 2Du - c_{\text{tot}}(0)]^2 \quad (15)$$

That is,

$$c_{\text{tot}}(0) = 1 + Du - Du \xi / [8R(1+Du)] \quad (16)$$

$$c_{\text{tot}}(1) = 1 + Du \xi / [8R(1+Du)] \quad (17)$$

The resistivity denominator reads:

$$R = \int dx / c_{\text{tot}} = 4R / Du \xi [c_{\text{tot}}(1) - c_{\text{tot}}(0)] = 1/(1+Du) \quad (18)$$

in which the Dukhin number corresponds to the surface transport contribution to the conductance. Thus,

$$c_{\text{tot}}(x) = [(1 - Du \xi/8)^2 + (1+Du) Du \xi/2 x]^{1/2} \quad (19)$$

7.2.2 Osmotic pressure

Finally, the average density reads:

$$\langle c_{\text{tot}} \rangle = 4 (c_{\text{tot}}(1)^3 - c_{\text{tot}}(0)^3) / [3Du \xi(1+Du)] \quad (20)$$

that is

$$\langle c_{\text{tot}} \rangle = 4/3 Du \xi(1+Du) [3Du\xi(1+Du)^2/4 + 2Du^3 \xi^3/8^3] = 1+Du+Du^2 \xi^2/[192(1+Du)] \quad (21)$$

meaning that we have an over-concentration inside the channel, corresponding to an average osmotic pressure of:

$$\Delta\Pi = 2c_0T (\langle c_{\text{tot}} \rangle - 1 - Du) = (c_0T Du^2 \Delta V^2) / (96(1+Du)T^2) = \Sigma^2 \Delta V^2 / [96(1+Du) h^2 c_0 k_B T] \quad (22)$$

where we have reintroduced the dimensions in the last expressions. Note that the pressure is positive independently of the sign of ΔV .

Using Eq. (7), this model predicts that the height or contrast of such a blister in its receding dynamics scales with $\Delta V^{2/3}$, which is consistent with experimental data (see Supplementary Fig. 24a).

7.3. Unidirectional memristors: Electrostatic pressure

7.3.1 Capacitive charging

First of all, let us check how the capacitive effect compares with the surface charge. We note that the top layer has capacitance around:

$$C_0 \approx \epsilon_0 \epsilon_{\text{hBN}} / t_L \approx 0.5 \text{ mF/m}^2 \quad (23)$$

The capacitive charging due to the voltage drop can then be estimated as:

$$\Delta\Sigma = C_0 \Delta V \approx 0.5 \text{ mC/m}^2 \quad (24)$$

which is smaller than the typical surface charge on hBN. Thus, we can neglect any effect related to capacitive charging.

7.3.2 Debye double-layer

The Debye double-layer adds a capacitance C_D in parallel to the hBN layer on each side, thus reducing the voltage drop experienced by the solid:

$$\Delta V_{\text{eff}} \approx \Delta V / (1 + 4C_0/C_D) \quad (25)$$

We can estimate the ratio of capacitance as:

$$C_0/C_D \approx \lambda_D/t_L \times (\epsilon_{\text{hBN}}/\epsilon_w) \approx 10^{-3} \quad (26)$$

so that the Debye's capacitance is much larger and then short-circuited by the solid's capacitance. Therefore, we can neglect the capacitive contribution from the Debye double-layer.

7.3.3 Electrostatic pressure due to surface charge

The surface charge will experience the local electric field and then induce a pressure force on the solid. Since the surface charges we consider are in the first layer close to the interface, they are screened by water in the parallel direction but mostly not in the normal direction⁸. Therefore, the normal contribution of the electric field at the surface mostly fully drags the surface charges. The situation is quite different for the counter charges which are further from the interface and therefore screened by water in every direction. These countercharges thus experience a much smaller local electric field in the normal direction. This justifies focusing only on the effect on the surface charges of the normal component of the electric field. Due to the geometry of the system and given that we can neglect of the capacitive effect of the Debye double-layer, the normal electric field going throughout the top layer is estimated close to the center as:

$$E_{\perp} \approx \Delta V / (\epsilon_{\perp_s} t_L) \quad (27)$$

where $\epsilon_{\perp_s} \approx 2.1$ according to ref 8. Thus, the force applied on the surface charge can be estimated as:

$$P_{\text{elec}} = 2\Sigma E_{\perp} \approx 2\Sigma \Delta V / (\epsilon_{\perp_s} t_L) \quad (28)$$

which should be enough to reach the threshold to form a blister.

Using Eq. (7), this model predicts that the height or contrast of such a blister in its receding dynamics scales with $\Delta V^{1/3}$, which is consistent with experimental data (see Supplementary Fig. 24a).

7.4. Dependence on salt concentration

Due to surface charge regulation, the surface charge is expected to depend on the salt concentration⁹. While this dependence adds complexity, we assume a standard scaling $\Sigma \sim c_0^{1/3}$ (see Supplementary Fig. 24b). Assuming such a scaling, we can study the maximum blister height at 1 V as a function of the salt concentration: the results are given in Supplementary Fig. 24c-d. We recover the experimental insight that the effect disappears at small concentrations (Supplementary Section 7.2). For the electrostatic pressure (Supplementary Fig. 24d), at lower

salt concentration, the surface charge is smaller and no longer reaches the adhesion threshold between 50 and 500 mM.

For the osmotic pressure (Supplementary Fig. 24c), the effect is more subtle because the selectivity also depends on the salt concentration: at large concentration there is a small Dukhin number and then a low selectivity. For our model to be tractable, we have assumed a strong polarisation effect in order to neglect the total salt current. This approximation becomes invalid at low selectivity, that is above 200 mM ($Du \sim 1$). Below this salt concentration, the osmotic pressure is predicted to scale with the surface charge and then to increase with the salt concentration. For larger salt concentrations, our model is no longer valid but we do expect a saturation. Thus, we can expect the extrapolation of our model up to 1 M to remain qualitatively acceptable.

Supplementary Discussion. Evidence that electromechanical hysteresis overrides purely ionic contributions to memory

In this work, our operando optical interferometry directly measures nanoscale structural deformations rather than local ion concentrations. Based on these observations and in particular, the symmetry of the voltage response, we infer that ionic concentration polarization (CP) likely provides the osmotic pressure responsible for bidirectional blister actuation. Nevertheless, we have strong evidence that purely ionic effects cannot account for the memristive hysteresis observed in these 2D nanochannels. Attributing the observed conductance memory solely to ionic microstates leads to fundamental physical contradictions. Instead, our measurements and theoretical boundary calculations establish that macroscopic structural deformation acts as the primary state variable governing the memory effect. This conclusion is supported by three independent lines of direct evidence:

1. Static vs. dynamic geometry: the absence of memory without deformation

The necessity of mechanical deformation is evidenced by the absence of memory when the channel geometry remains static. In highly confined regimes where CP is active, the device response remains linear if the van der Waals slit does not actively deform (Main text Fig. 2a). This is true for higher voltage ranges where devices show no active blister deformation that are contributing to conductance of the device (OFF-state conductance at negative voltages in Fig. 4d, fully linear I - V curves in Supplementary Fig. 9b, negative and positive parts of the I - V curves in Supplementary Fig. 20a and 20c, respectively). Hysteresis only emerges in precise spatiotemporal synchrony with dynamic blister growth and relaxation. Furthermore, control devices operating in regimes of negligible ionic selectivity (e.g., $h \approx 50$ nm slits) show clear blister nucleation driven by electrostatic forces without exhibiting macroscopic I - V hysteresis (Supplementary Fig. 11c), demonstrating that blisters can form independently of CP.

2. Baseline delamination and conductance limits

To quantify the geometric contribution to the observed memory, we calculate the theoretical conductance of a perfectly flat, rigid van der Waals nanoslit. For a standard

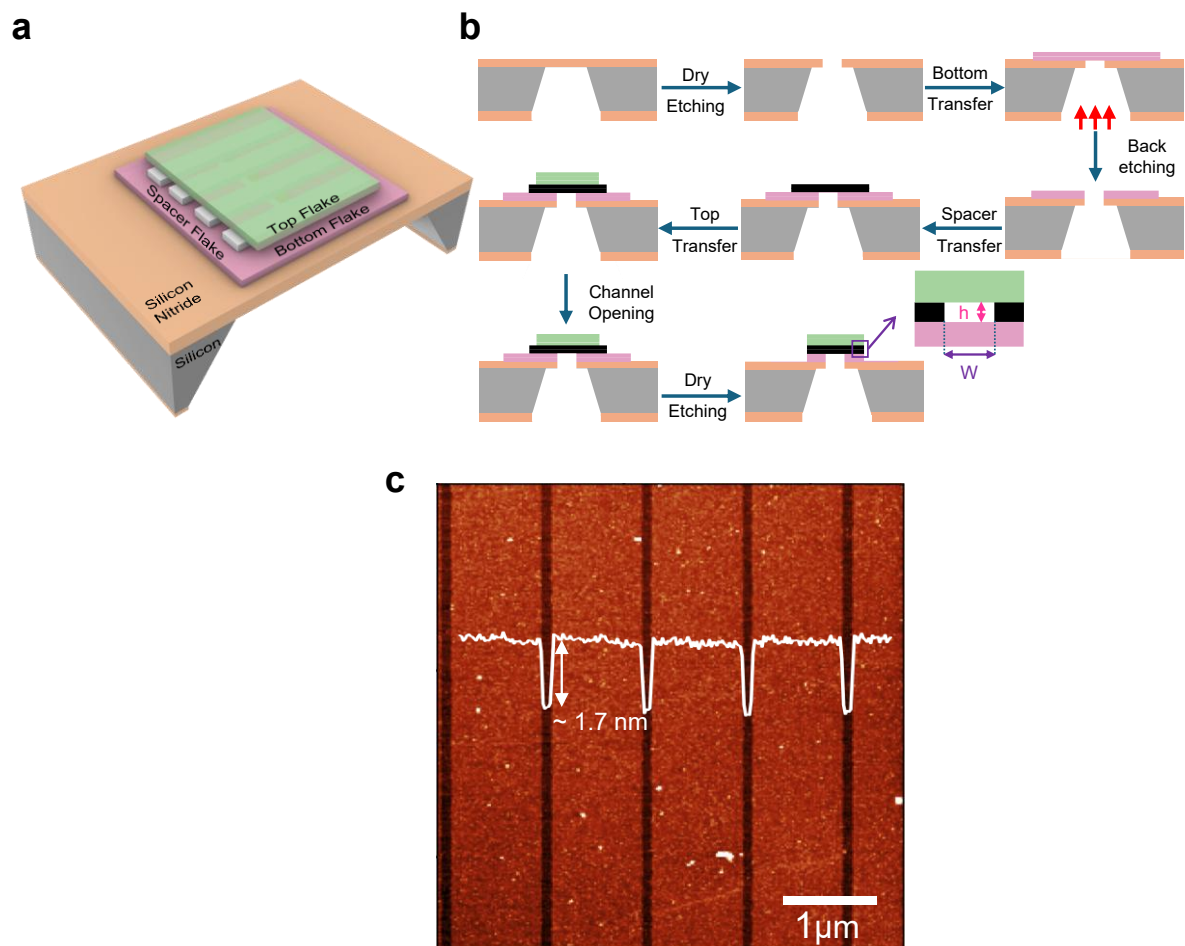
device geometry ($N=60$, $L \approx 4 \text{ } \mu\text{m}$, $w=100 \text{ nm}$, and $h=0.7 \text{ nm}$) filled with 1 M KCl (conductivity $\sigma \approx 10 \text{ S/m}$), the baseline conductance is defined as: $G_{flat} = N \cdot \sigma \cdot w \cdot h / L \approx 10.5 \text{ nS}$. The experimentally measured baseline of $\sim 40 \text{ nS}$ at 0 V confirms that the channel is inherently non-rigid and already partially delaminated prior to out-of-plane biasing.

3. Physical limits of ionic memory

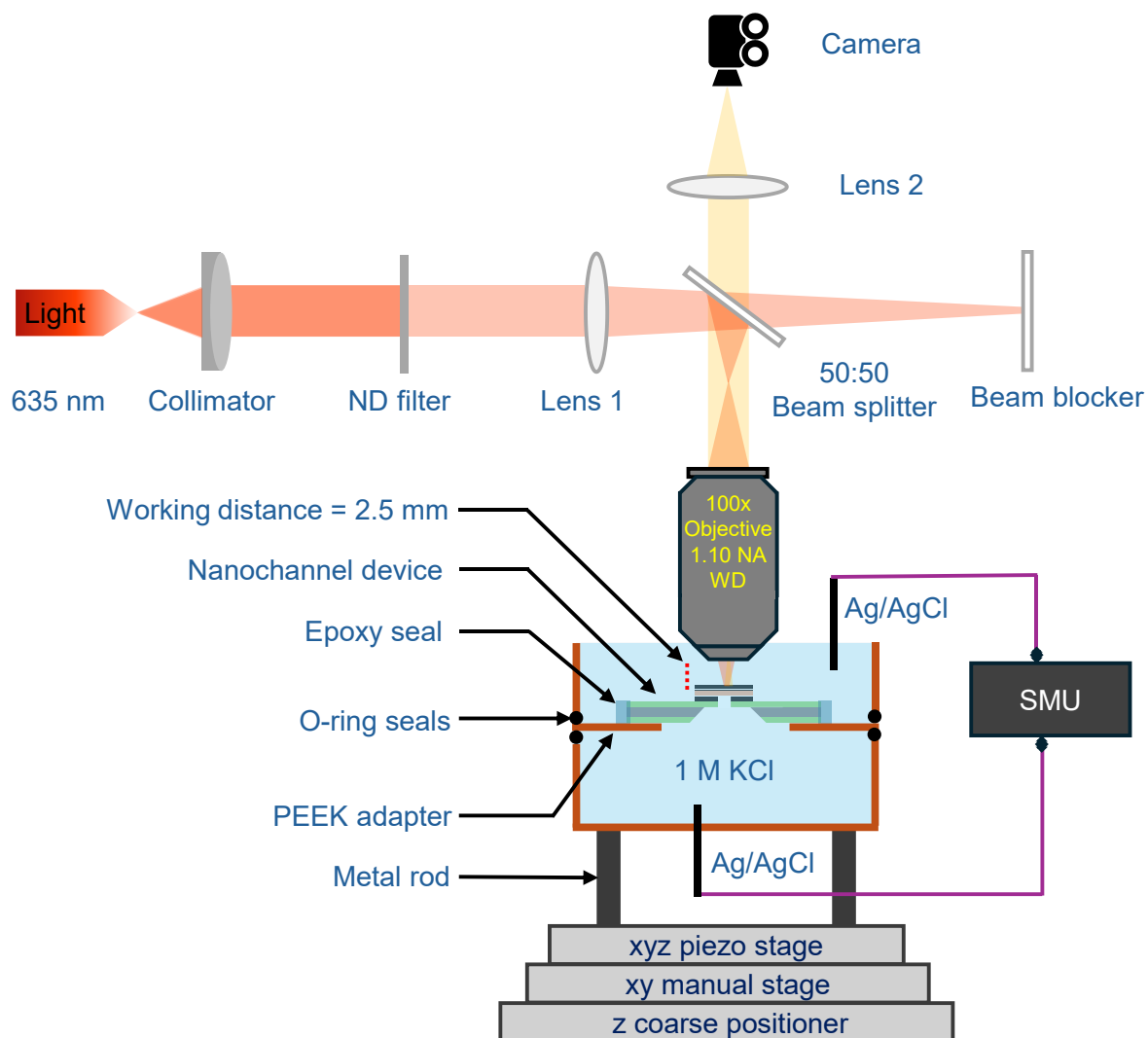
During dynamic operation, the memristive switching exhibits discrete conductance jumps. When a characteristic dynamic blister forms (e.g., with an estimated height $h_B \approx 10 \text{ nm}$), its structural contribution to the channel cross-section can be approximated as $G_{blister} \approx \sigma \cdot h_B \approx 100 \text{ nS}$. This purely geometric deformation observation perfectly accounts for the measured $\sim 100 \text{ nS}$ discrete conductance jumps.

Conversely, attributing these massive conductance modulations solely to ionic changes is physically impossible. The devices routinely exhibit ON/OFF conductance ratios of approximately 50 (Main Text, Fig. 3c-d). Generating a 50-fold conductance increase in a rigid 0.7 nm channel via ionic effects alone would require the local KCl concentration to surge from 1 M to 50 M. Because the theoretical aqueous solubility limit of KCl is $\sim 4.8 \text{ M}$, such a concentration is unattainable. Therefore, while ionic pressure triggers the phenomenon, the macroscopic mechanical deformation of the compliant channel walls is the necessary physical state variable that enables the memristive hysteresis.

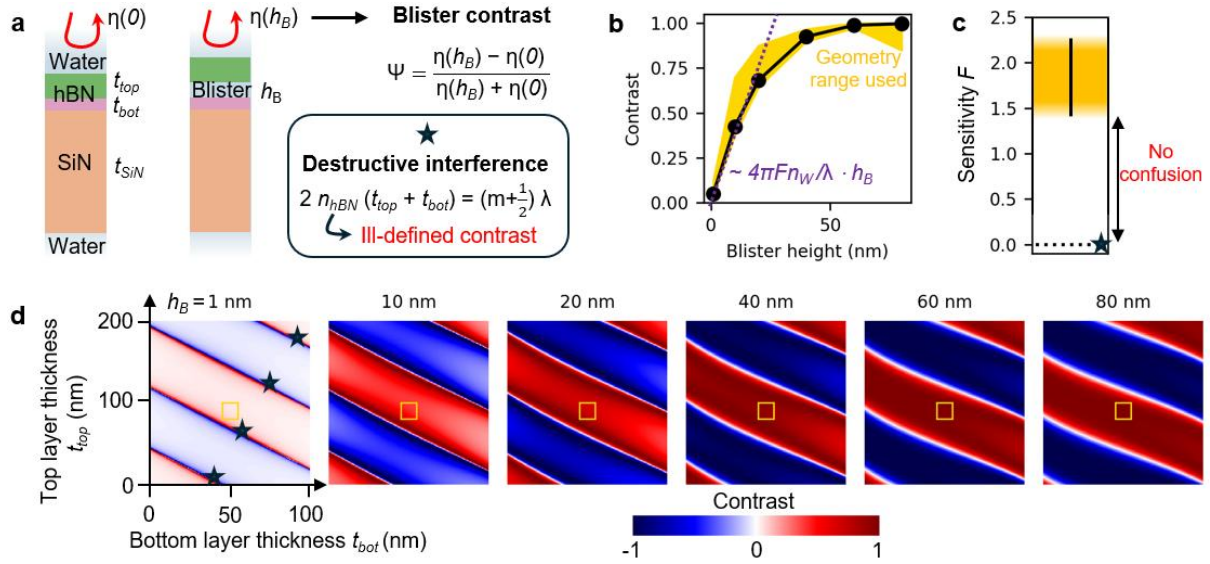
Supplementary Figures 1 - 24



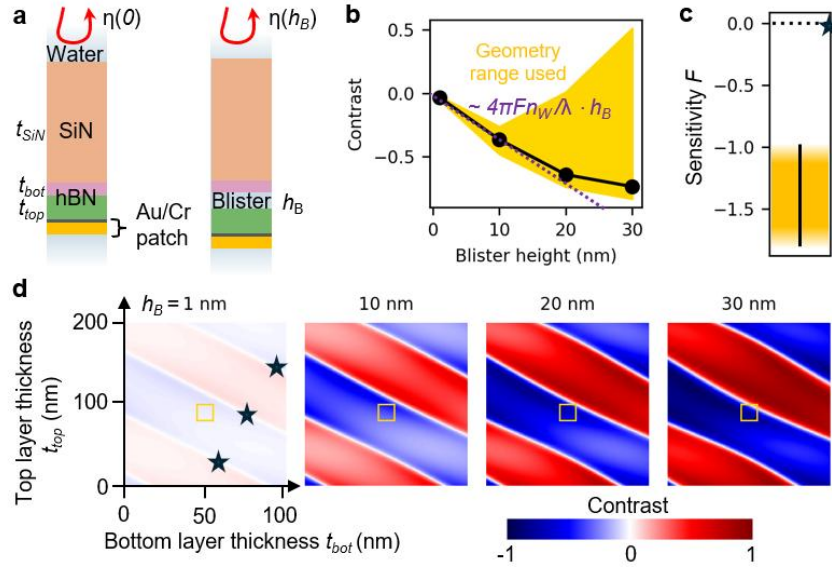
Supplementary Fig. 1 | (a) Schematic of a nanochannel device (b) Schematic of the fabrication step involved. A micro hole is made on the free-standing SiN_x membrane by photolithography, followed by dry etching. Onto this substrate, a hBN bottom flake is transferred and back etched (dry etching). Subsequently, a top flake is transferred above the spacer. A spacer flake is made by the e-beam lithography technique, wherein a parallel array of strips is patterned, followed by oxygen plasma etching. Then the top-spacer stack is further transferred onto the bottom substrate. To define channel length, a rectangular aperture is made using photolithography, followed by dry etching. (c) AFM profile of a five-layer graphene spacer flake with a channel width of ~ 100 nm separated by $\sim 1\mu\text{m}$.



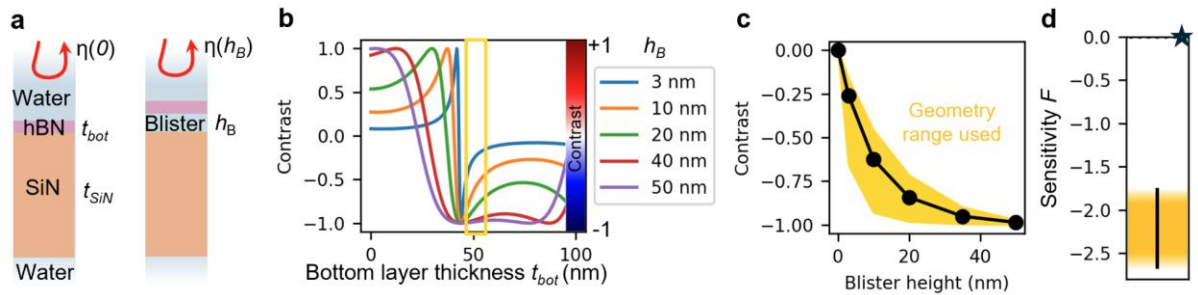
Supplementary Fig. 2 | Schematic of the setup. The setup consists of a white light source with a 10 nm bandpass filter with a central wavelength of 635 nm, focused on the back focal plane of a water-dipping objective, and further to the device immersed in an aqueous solution. The reflectance image is then acquired using a CMOS camera.



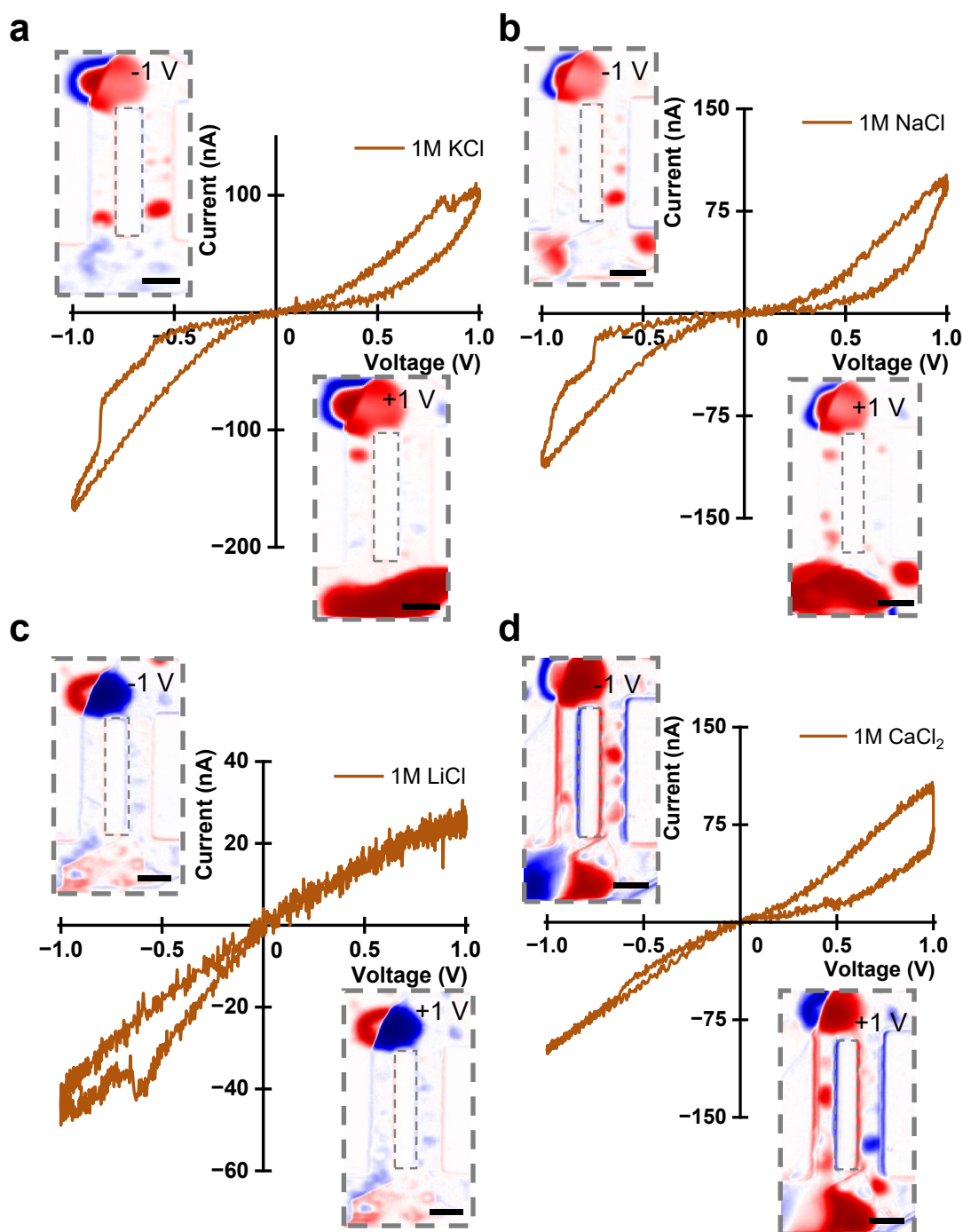
Supplementary Fig. 3 | Transfer matrix method (TMM) calculations of the optical contrast in the used device geometries. **a**, Sketch of the thin film structure at rest and in the presence of a blister, used to compute the VIC Ψ . In the pathological case of destructive interference, the contrast is ill-defined, which necessitates carefully chosen experimental parameters. **b**, Contrast vs blister height curve showing the derivation of the sensitivity from the near-zero slope. **c**, Sensitivity values extracted from the used geometry range show that the sensitivity is always a positive number exceeding 1.4, thereby providing unambiguous information about small deformations. **d**, TMM contrast map for varying bottom and top layer thicknesses, given for blister heights ranging from 1 nm to 80 nm. The parameters used correspond to the yellow box in which the formation of a blister always results in a positive contrast change in red. The presence of singularities in the contrast along tilted lines corresponds to the destructive interference condition in **a**, leading to ill-defined contrast and sensitivity.



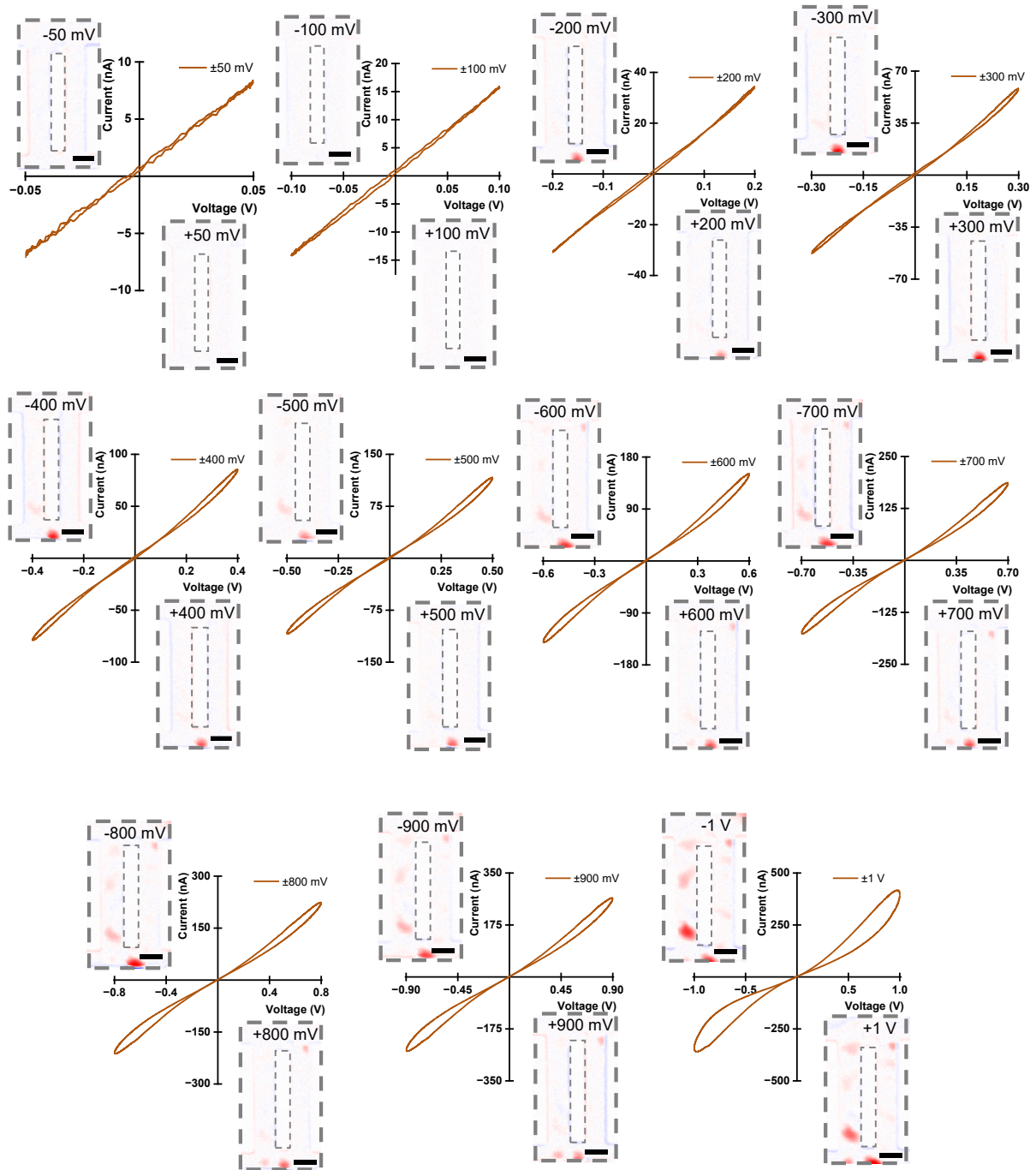
Supplementary Fig. 4 | Transfer matrix calculations of the optical contrast for gold patch devices imaged upside down. **a**, Sketch of the thin film structure at rest and in the presence of a blister, for a device placed upside down and including a gold/chromium patch. **b**, Contrast vs blister height curve showing negative sensitivity. **c**, Sensitivity values extracted from the used geometry range show that the sensitivity is always a negative number below -1.0, thereby providing unambiguous information about small deformations. **d**, TMM contrast map for varying bottom and top layer thicknesses, given for blister heights ranging from 1 nm to 30 nm. The parameters used correspond to the yellow box in which the formation of a blister always results in a negative contrast change in blue, for small enough deformations.



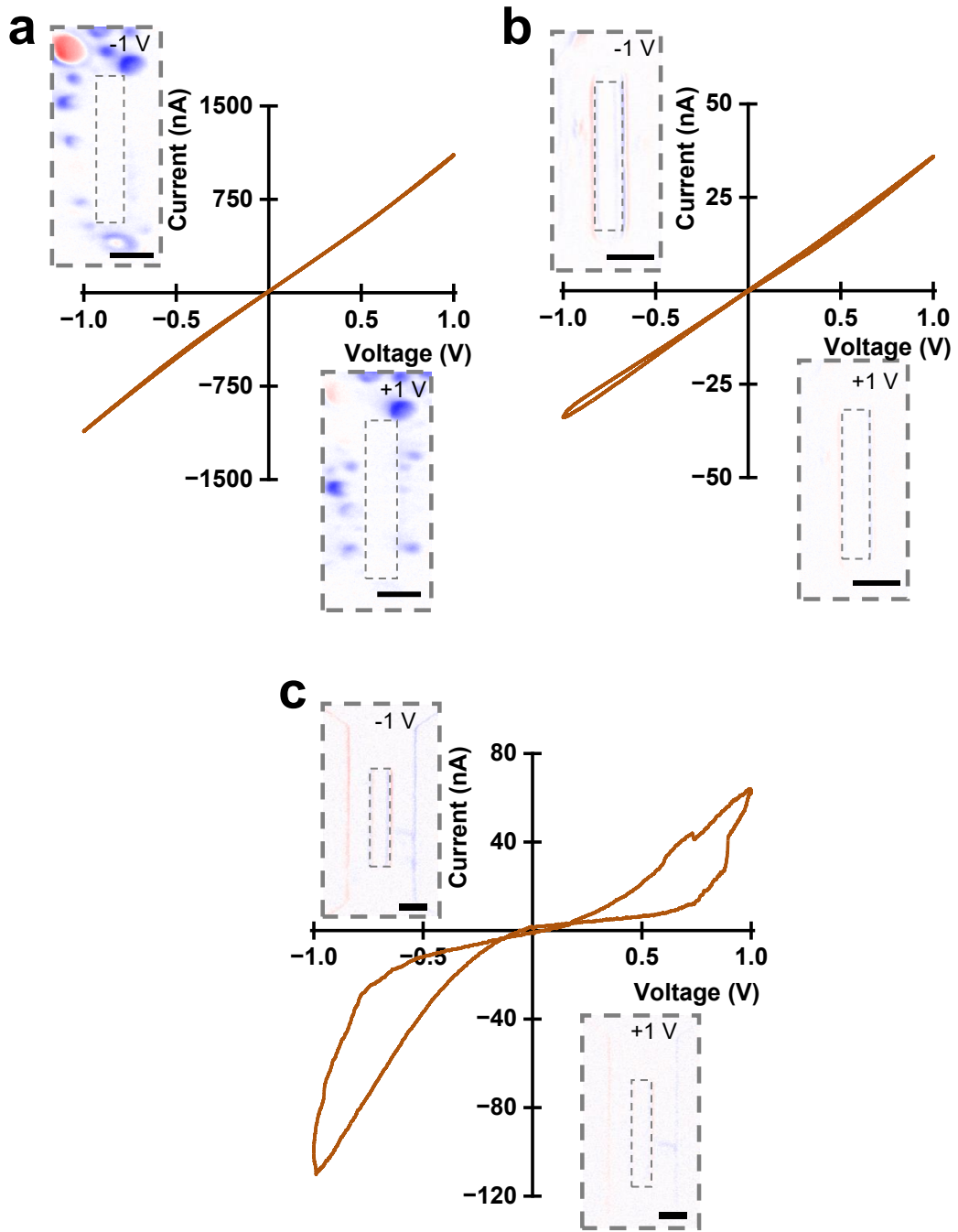
Supplementary Fig. 5 | Transfer matrix calculations of the optical contrast for topless parts of the devices. **a**, Sketch of the thin film structure at rest and in the presence of a blister forming between the bottom hBN and the SiN_x substrate. **b**, Contrast vs bottom layer thickness curve for different blister heights. **c**, Contrast vs height curve showing negative sensitivity for geometries used in this work. **d**, Sensitivity values extracted from the used geometry range show that the sensitivity is always a negative number below -1.8, thereby providing unambiguous information about small deformations.



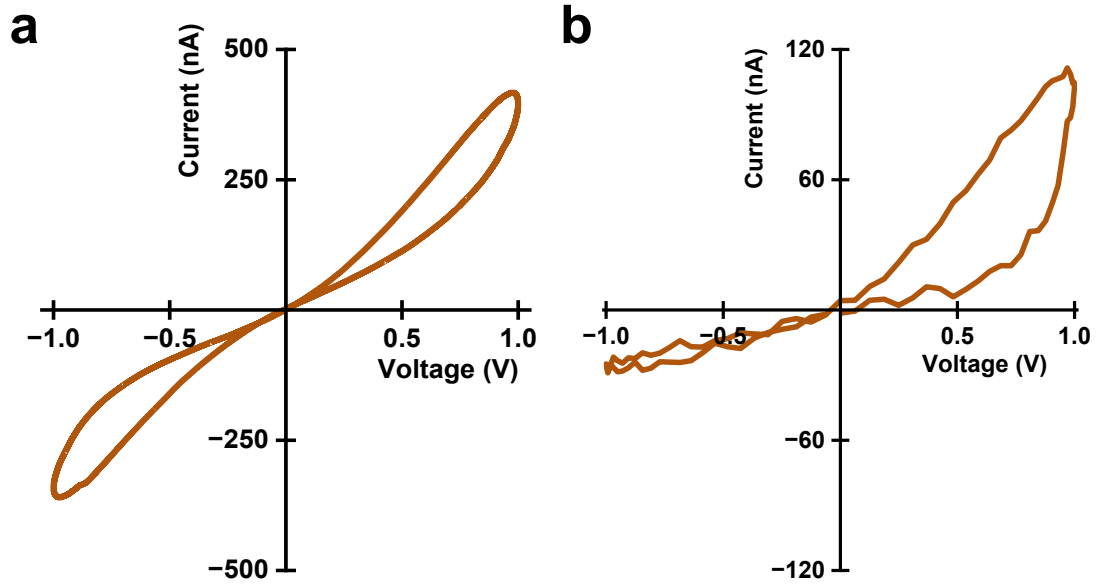
Supplementary Fig. 6 | *I-V* curves and VIC images of device 2 at ± 1 V with varying salt type (a) 1 M KCl, (b) 1 M NaCl, (c) 1 M LiCl and (d) 1 M CaCl₂ at a frequency $f = 10$ mHz. The *I-V* response and VIC differ across salt types with KCl and NaCl showing bidirectional hysteresis (current loops in both voltage polarity) and LiCl and CaCl₂ showing unidirectional hysteresis (current loops in either of the voltage polarity). Scale bar: 5 μ m



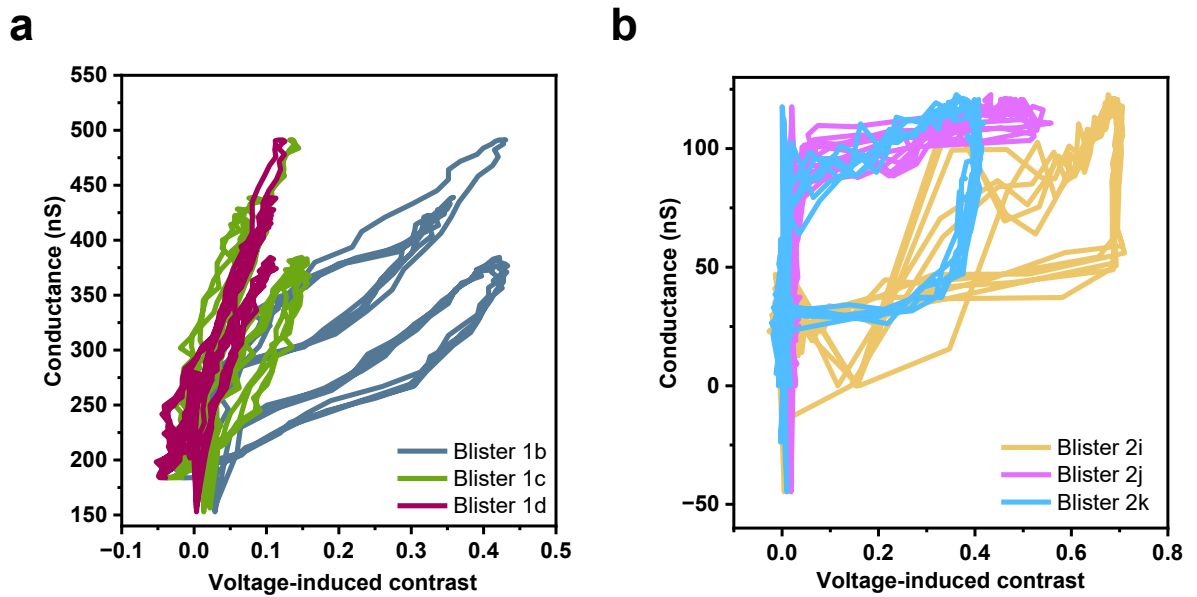
Supplementary Fig. 7 | Voltage Threshold. I - V curves and VIC images of device 1 at voltage extremes with varying potential range (50 mV-1 V) in 1 M KCl at a frequency $f=100$ mHz. Below threshold voltage ± 0.2 V, I - V is linear and the VIC images show no contrast. As the voltage amplitude increases beyond ± 0.2 V, the device shows hysteresis and VIC contrast appears and becomes more pronounced at higher voltage amplitudes. Scale bar: 5 μ m



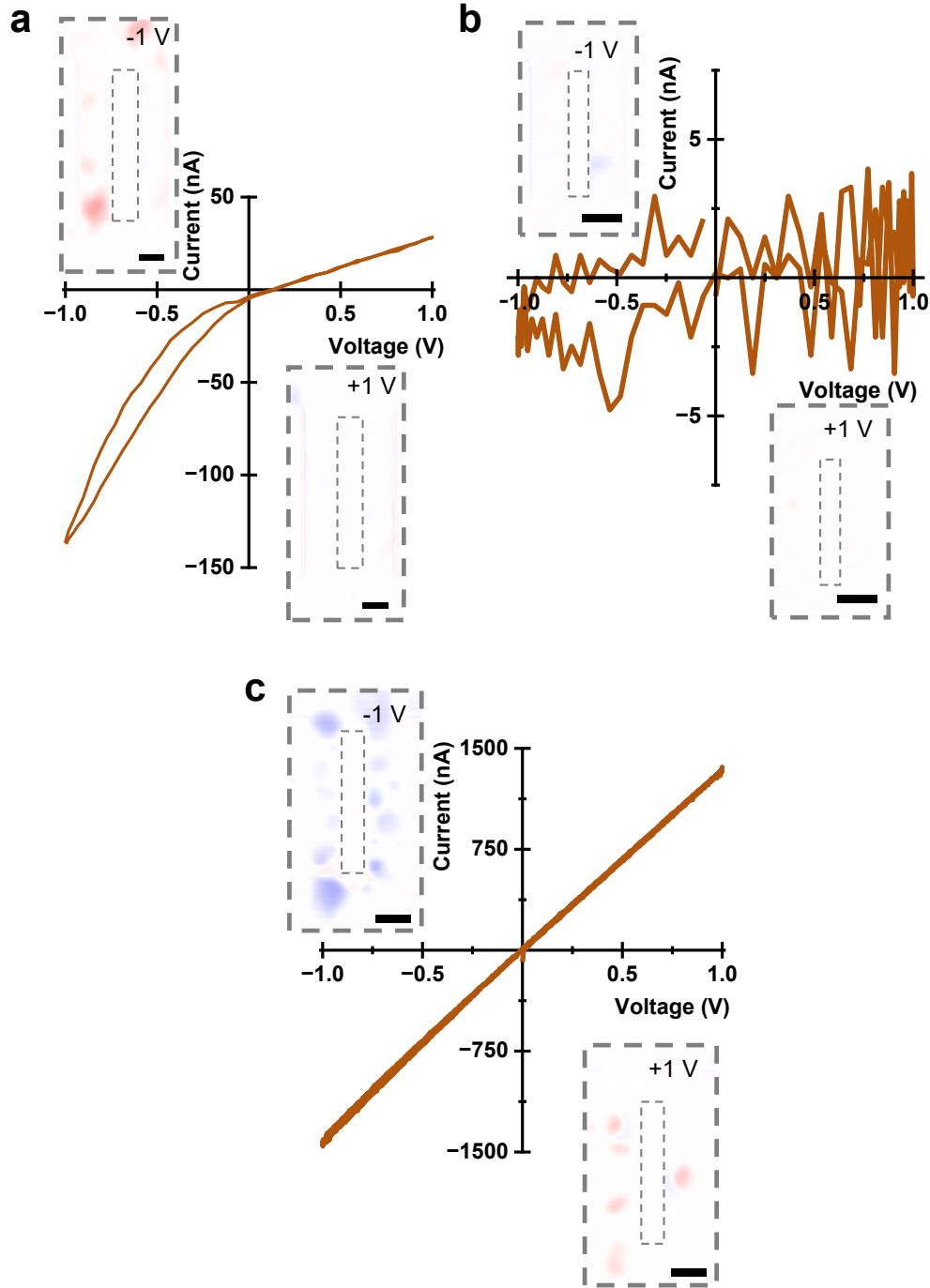
Supplementary Fig. 8 | I - V curves and VIC images at -1V and +1V of (a) device 12 (only bottom hBN, with no spacer or top) in 1 M KCl at a frequency $f = 10$ mHz. (b) device 13 (only bottom hBN, with no spacer or top) in 1 M KCl at a frequency $f = 10$ mHz and (c) device 5 (gold patch device) at -1 V and +1 V in 1 M KCl at a frequency of $f = 10$ mHz. (Here, the imaging is done from the bottom side of the device). Scale bar: 5 μ m



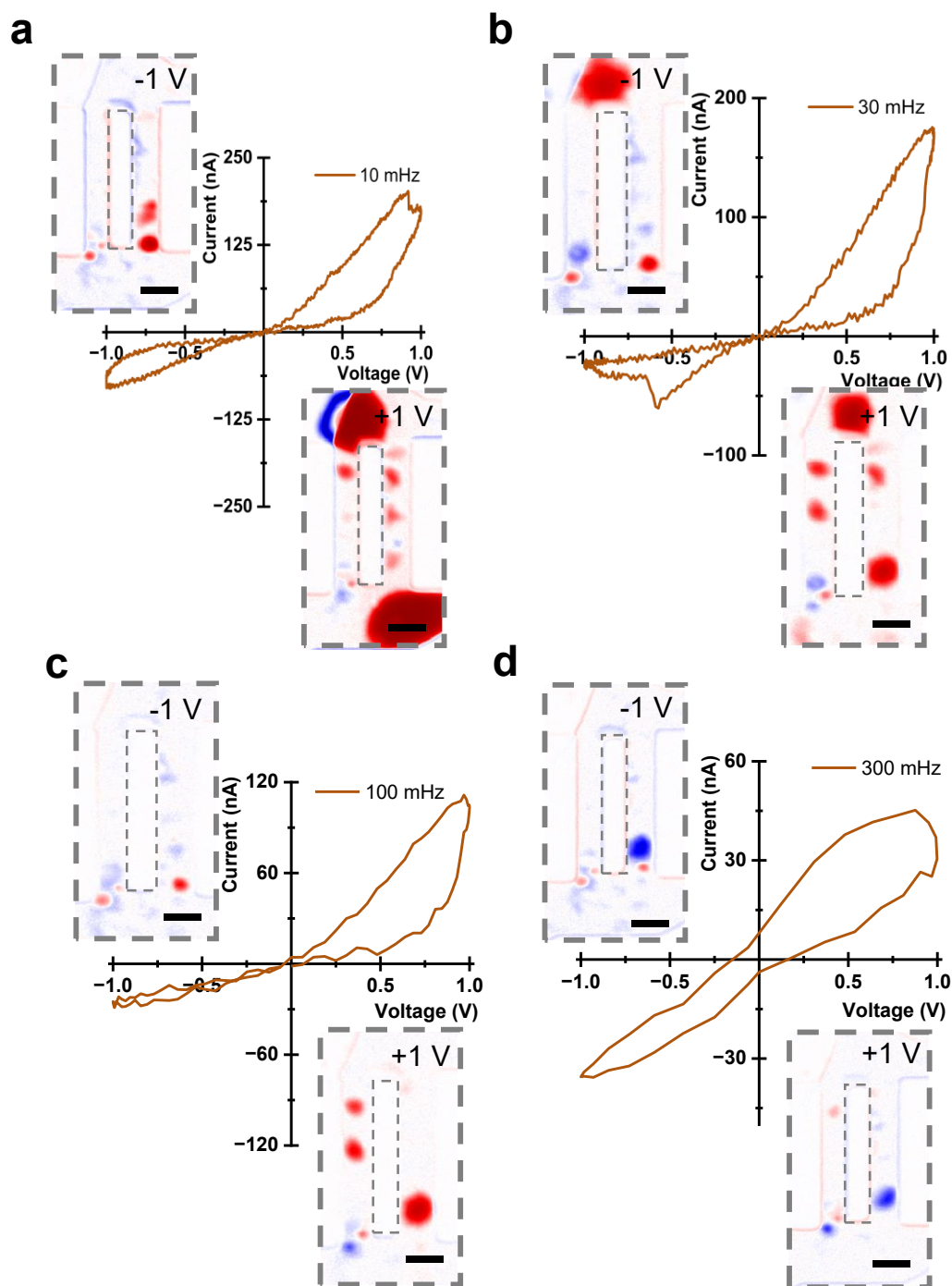
Supplementary Fig. 9 | I - V curves corresponding to VIC images presented in figures (a) 4a from device 1 and (b) 4b from device 2. Device 1 shows bidirectional hysteresis with current loops in both voltage polarity. Device 2 shows unidirectional hysteresis with loop confined to only positive voltage polarity.



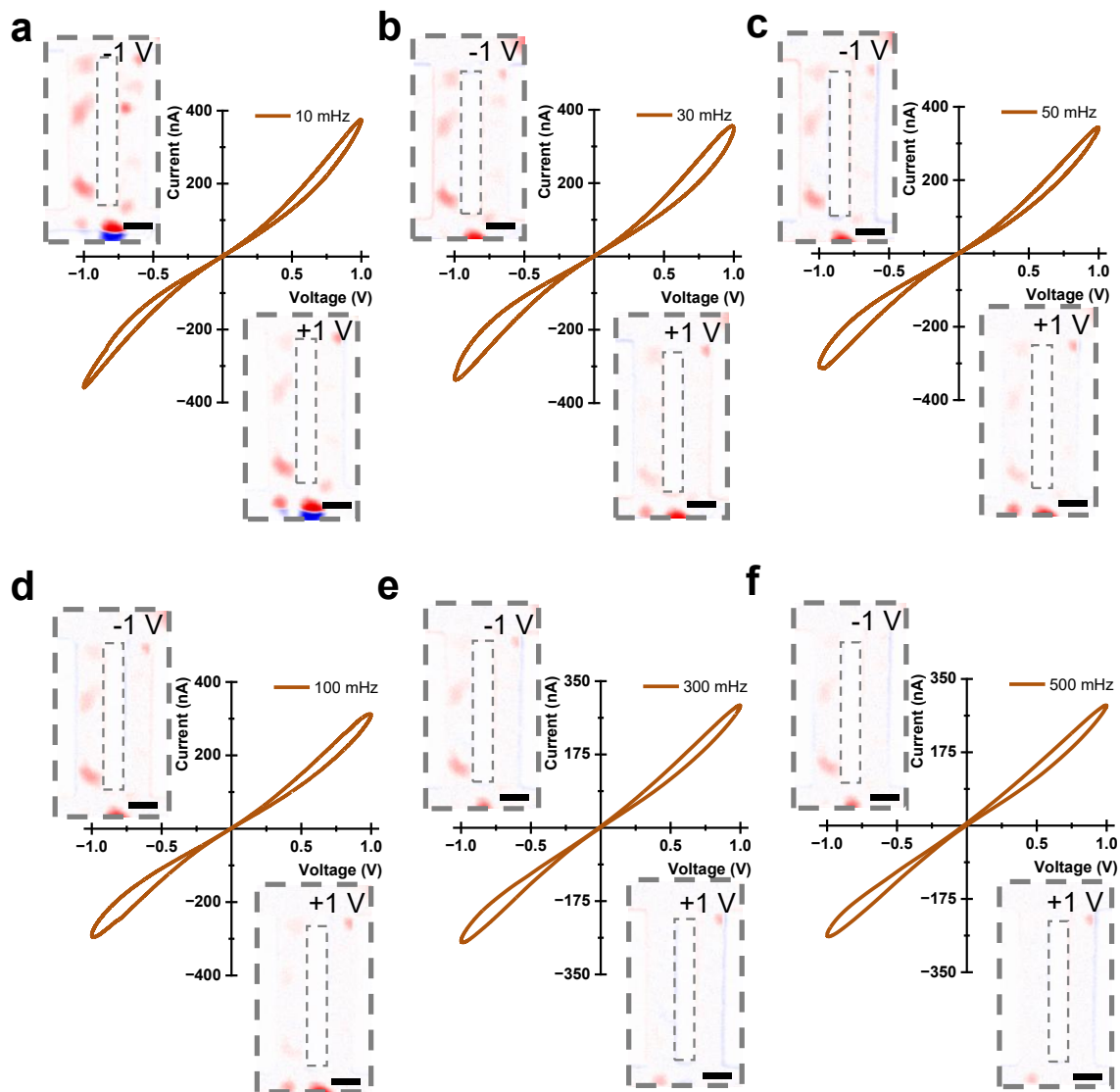
Supplementary Fig. 10 | Conductance vs voltage induced graph for the (a) bidirectional hysteresis memristive device 1 shown in figure 4a and (b) unidirectional hysteresis device 2 shown in figure 4b.



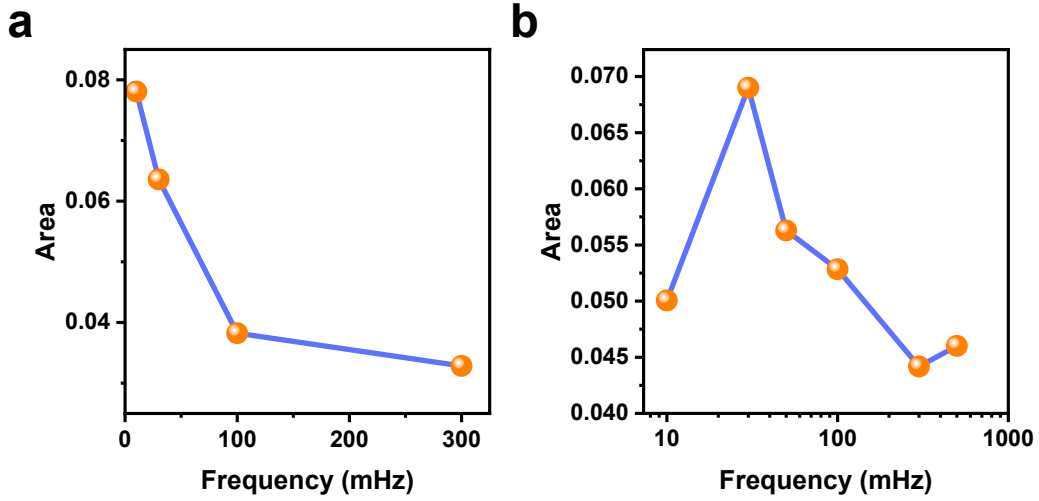
Supplementary Fig. 11 | *I-V* curves and corresponding VIC images at ± 1 V of (a) device 4 at -1 V and +1 V, showing blister responsiveness to negative voltage in 1 M KCl at a frequency $f = 100$ mHz. The device shows VIC change predominantly at negative voltage range. To test whether improved (dry state) surface cleanliness could reduce or suppress the blistering phenomenon, the device underwent annealing in 10% Hydrogen in Argon atmosphere at 300°C for 3 hours and 400°C for 4 hours. We observed that blisters could form despite annealing. (b) Device 11 (hBN as top and bottom and without any spacer) in 1 M KCl at a frequency $f = 100$ mHz with no VIC change at either voltage polarity and (c) device 7 (50 nm hBN spacer height) in 1 M KCl at a frequency $f = 10$ mHz showing VIC change at both voltage polarity ranges with no hysteresis. Scale bar: 5 μm



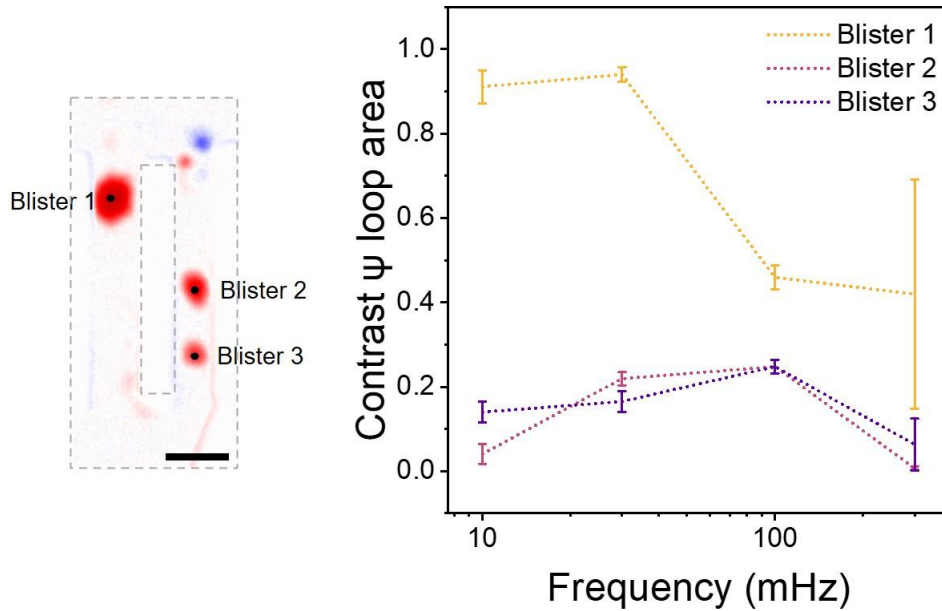
Supplementary Fig. 12 | I - V curves and VIC images of device 2 at ± 1 V with a varying frequency from 10 mHz to 300 mHz in 1 M KCl. The hysteresis transitions from bidirectional hysteresis to unidirectional at intermediate frequency and then becomes completely capacitive at 300 mHz with VIC diminishing across panels. Scale bar: 5 μ m



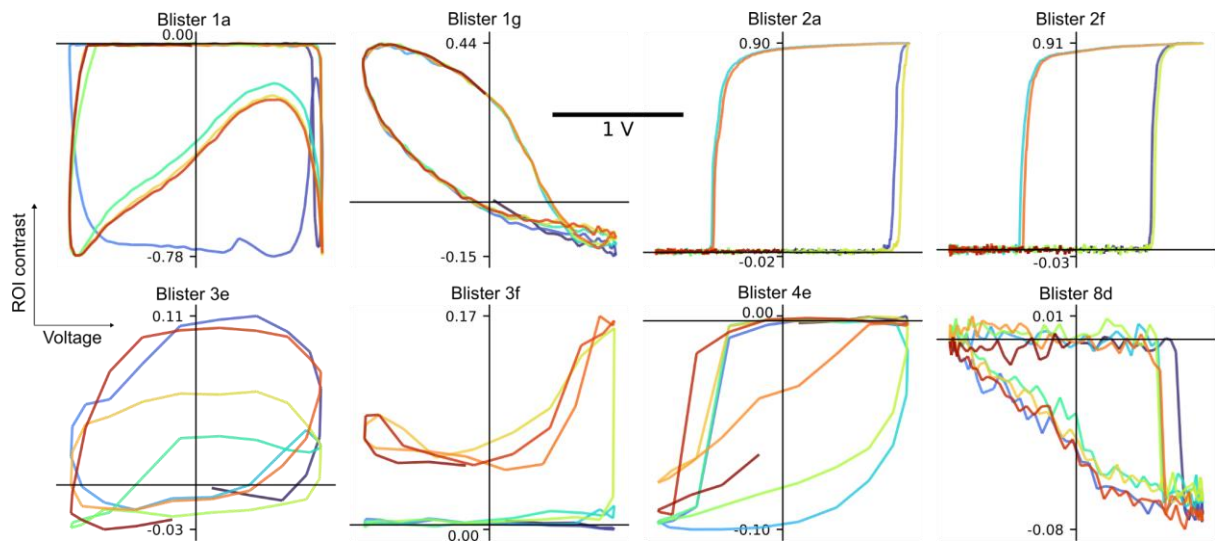
Supplementary Fig. 13 | I - V curves and VIC images of device 1 at ± 1 V with varying frequency (10 mHz to 500 mHz) in 1 M KCl. The bidirectional hysteresis loop reaches maximum at 30 mHz and narrows progressively with increasing frequency and the VIC diminishes correspondingly. The device remains bidirectional across the full frequency range. Scale bar: 5 μ m



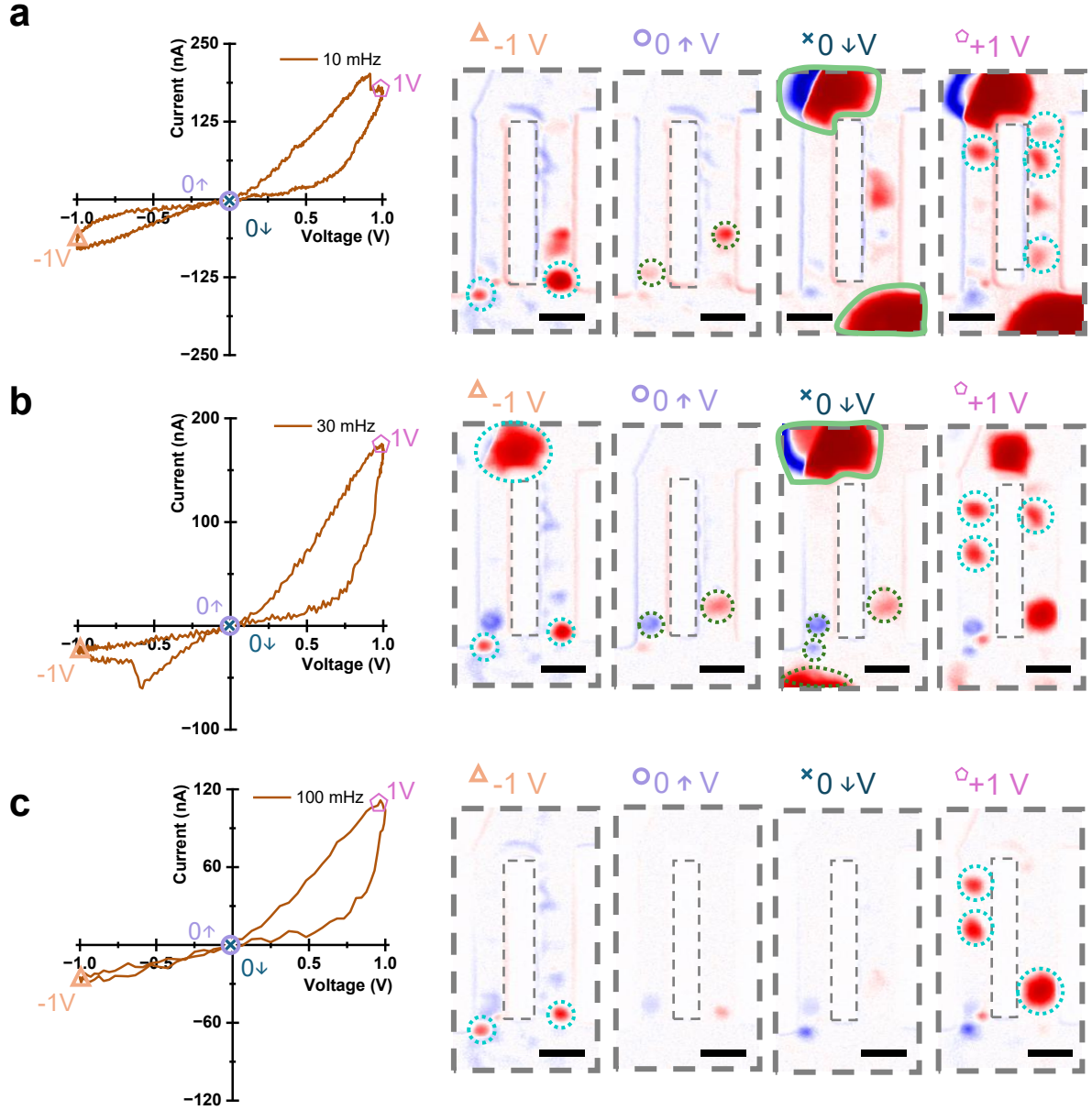
Supplementary Fig. 14 | Hysteresis loop area as a function of frequency for (a) Device 2 and (b) Device 1, measured in 1 M KCl. For device 2, the loop area decreases monotonically. Device 1 shows a peak loop area 30 mHz before declining at higher frequencies. The corresponding I - V curves and VIC images for device 2 and device 1 are shown in Supplementary Figure 12 and Supplementary Figure 13, respectively. In both the devices, the highest loop area is observed at lower frequencies.



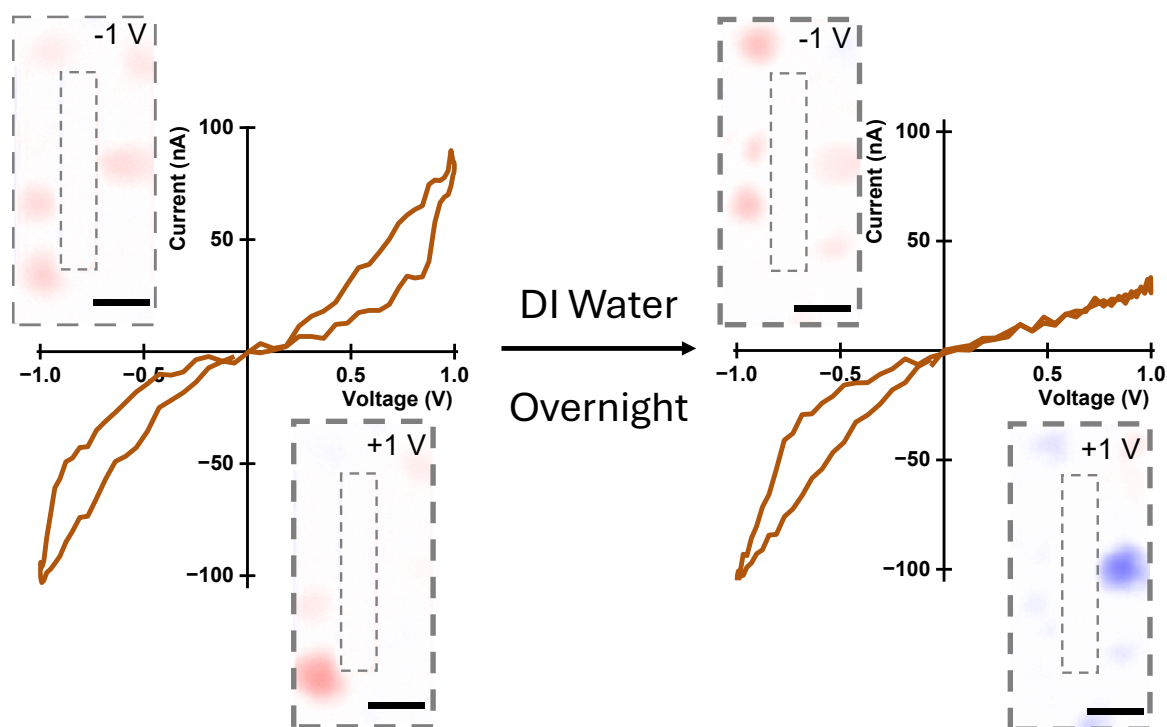
Supplementary Fig. 15 | Per-blister breakdown VIC loop area vs frequency for selected blisters of device 2, measured in 1 M KCl with error bar from multiple area cycles (For $f = 10$ mHz; $n = 2$ cycles, $f = 30$ mHz; $n = 2$ cycles, $f = 100$ mHz; $n = 6$ cycles, $f = 300$ mHz; $n = 6$ cycles). The left panel shows a VIC image of device 2 with three blisters labelled. For blister 1, the maximum contrast loop area is observed at 30 mHz whereas for blister 2 and 3, it occurs at 100 mHz. Scale bar: 5 μ m



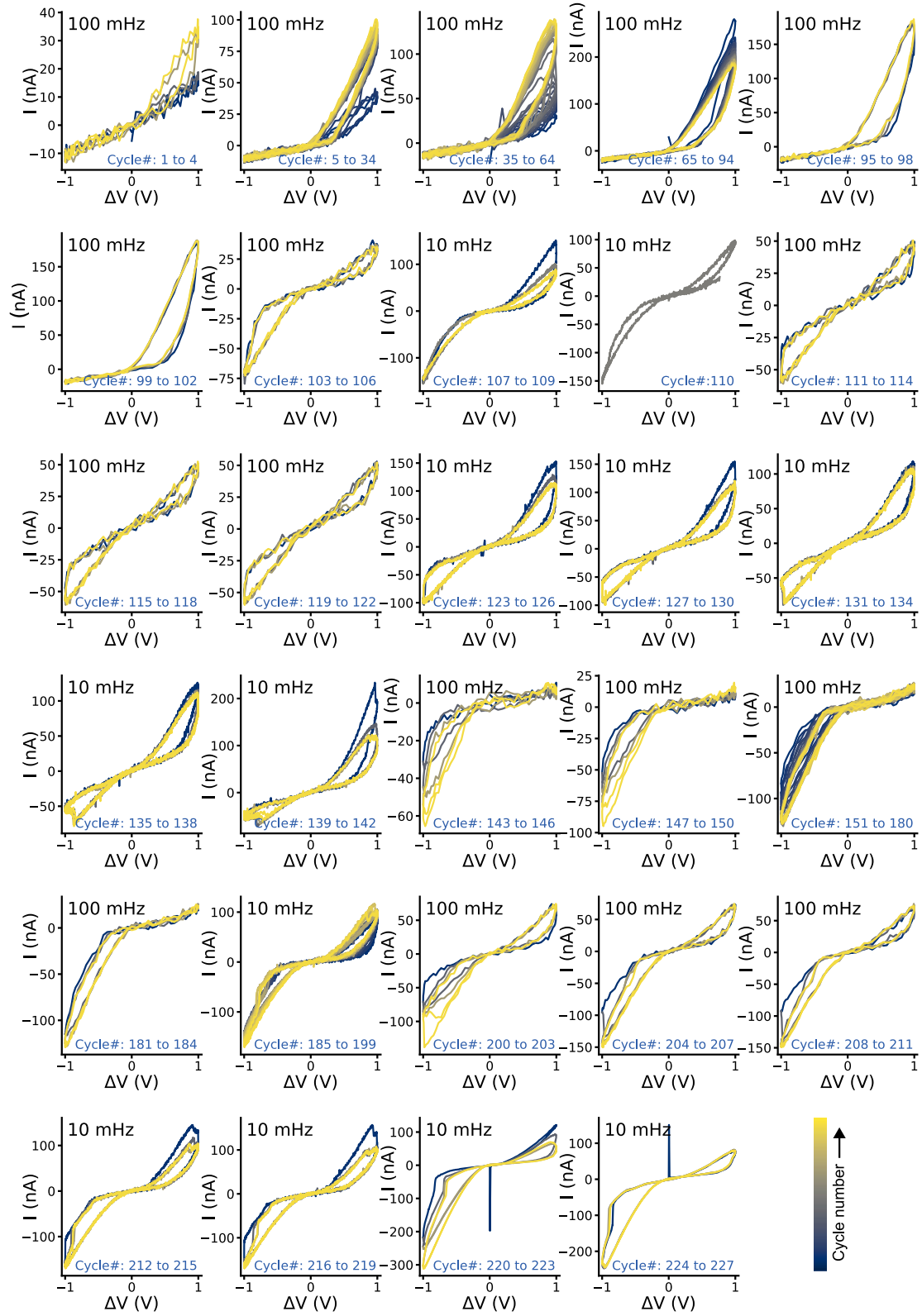
Supplementary Fig. 16 | Contrast-voltage curves of blisters exhibiting contrast retention at 0V, 8 occurrences out of 50 measurements. While the majority of blisters fully relax to their baseline state at 0 V (N=42, non-retentive), a specific subset (N=8, presented here) maintains a significant structural deformation upon voltage removal, providing the physical basis for non-volatile memory behaviour. We note that all such retentive blisters identified in our analysis are unidirectional.



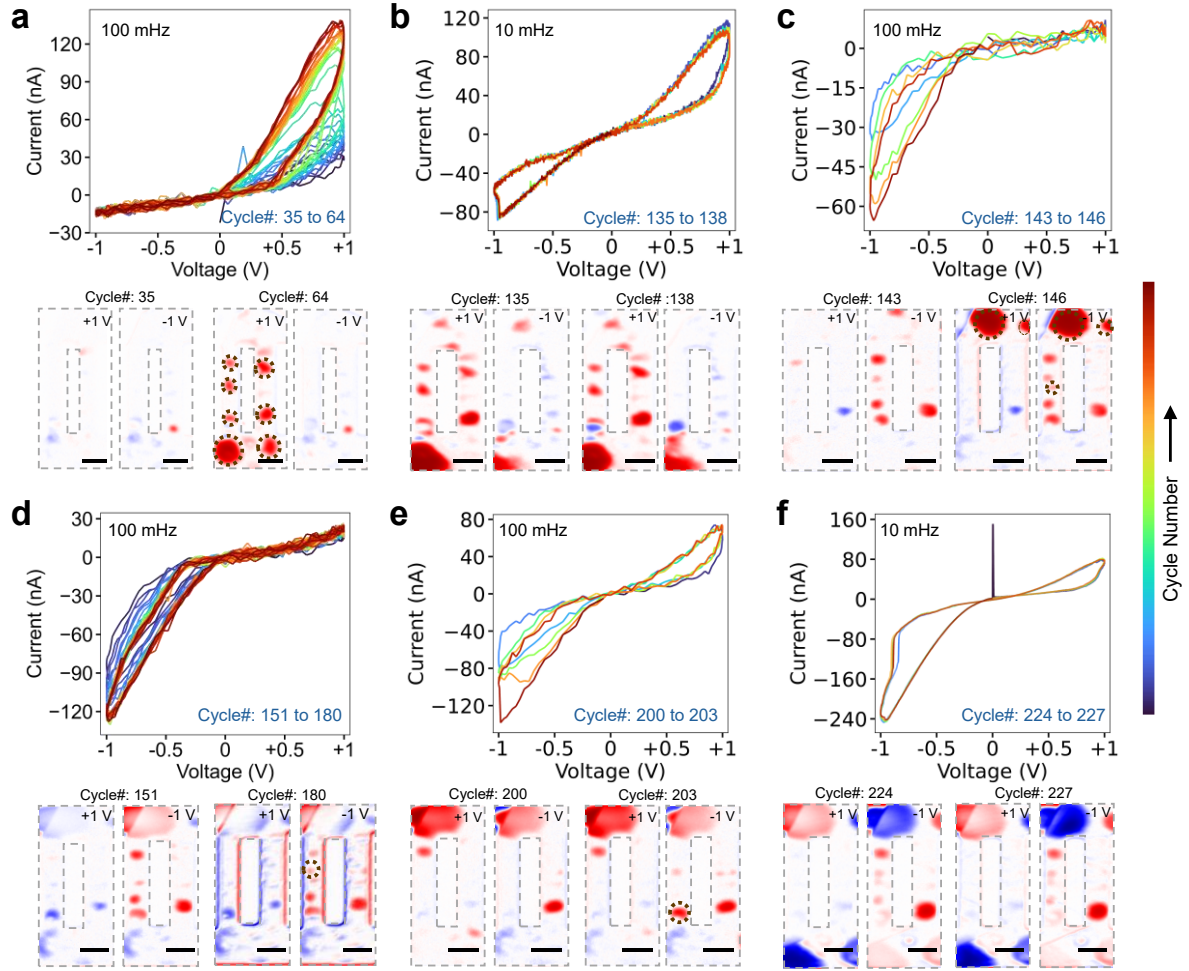
Supplementary Fig. 17 | Frequency-dependent transition between volatile and non-volatile structural states. **a–c**, Operando VIC images and synchronized I - V curves for device 2 at driving frequencies of **(a)** 10 mHz, **(b)** 30 mHz, and **(c)** 100 mHz. Images are captured at voltage extrema (± 1 V) and zero-crossings ($0 \downarrow$ V and $0 \uparrow$ V). At lower frequencies (10–30 mHz), large blisters (light green outlines) persist after bias removal, resulting in non-volatile memory. In contrast, smaller blisters (cyan outlines) collapse immediately upon bias removal. At 100 mHz **(c)**, the system transitions to a purely volatile regime where no retentive blisters are observed, directly corresponding to the suppression of the hysteretic state at the 0 V crossing in the electrical transport data. Scale bar: 5 μm .



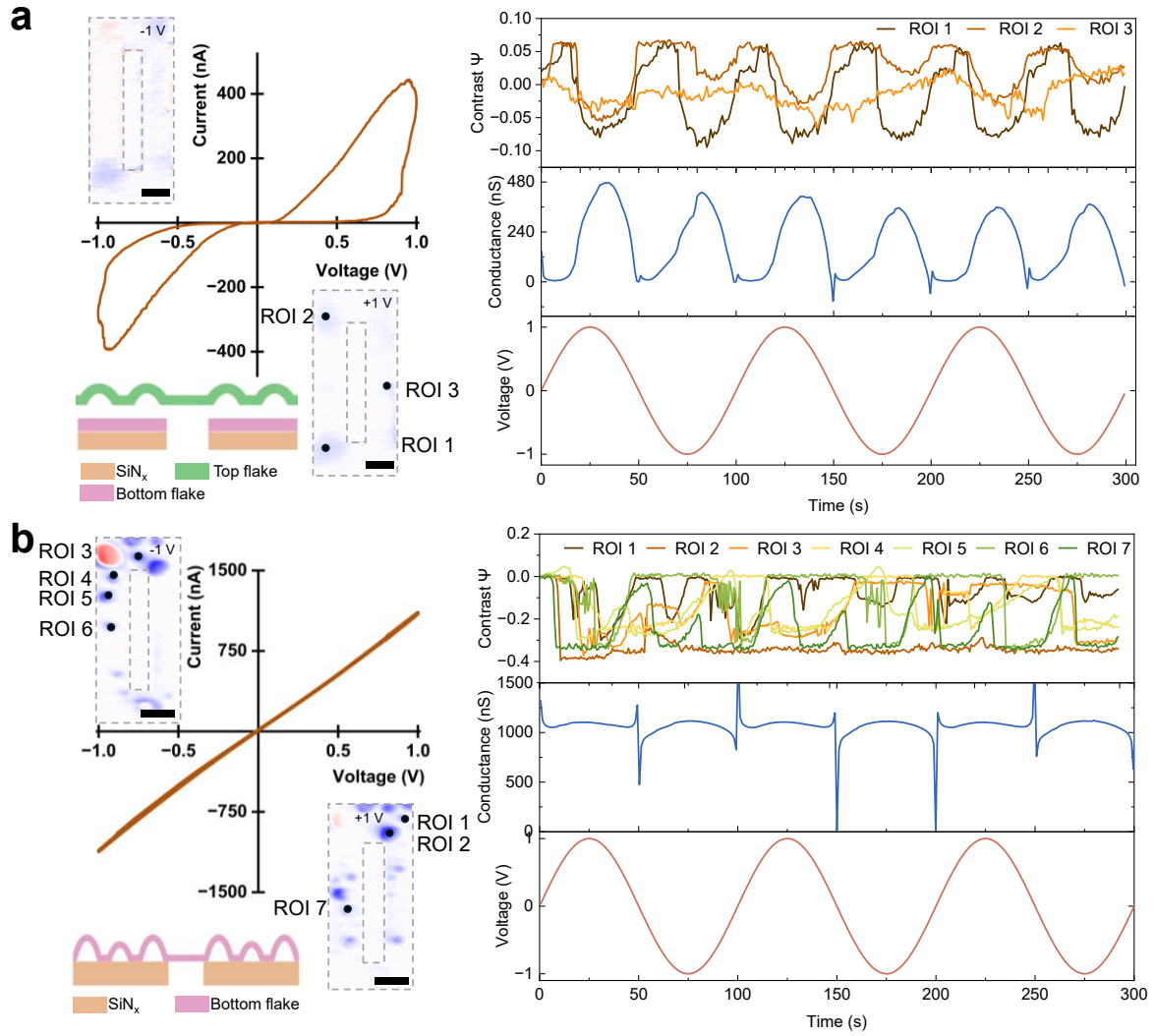
Supplementary Fig. 18 | I - V curves and VIC images of device 3 at ± 1 V in 1 M KCl at a frequency $f = 100$ mHz. The left panel shows the device before overnight submersion in DI water, where the I - V response exhibits bidirectional hysteresis. After leaving the device submerged in water overnight, the hysteresis shifts to unidirectional as shown in the right panel and VIC images at both the bias polarities change accordingly. Scale bar: 5 μm



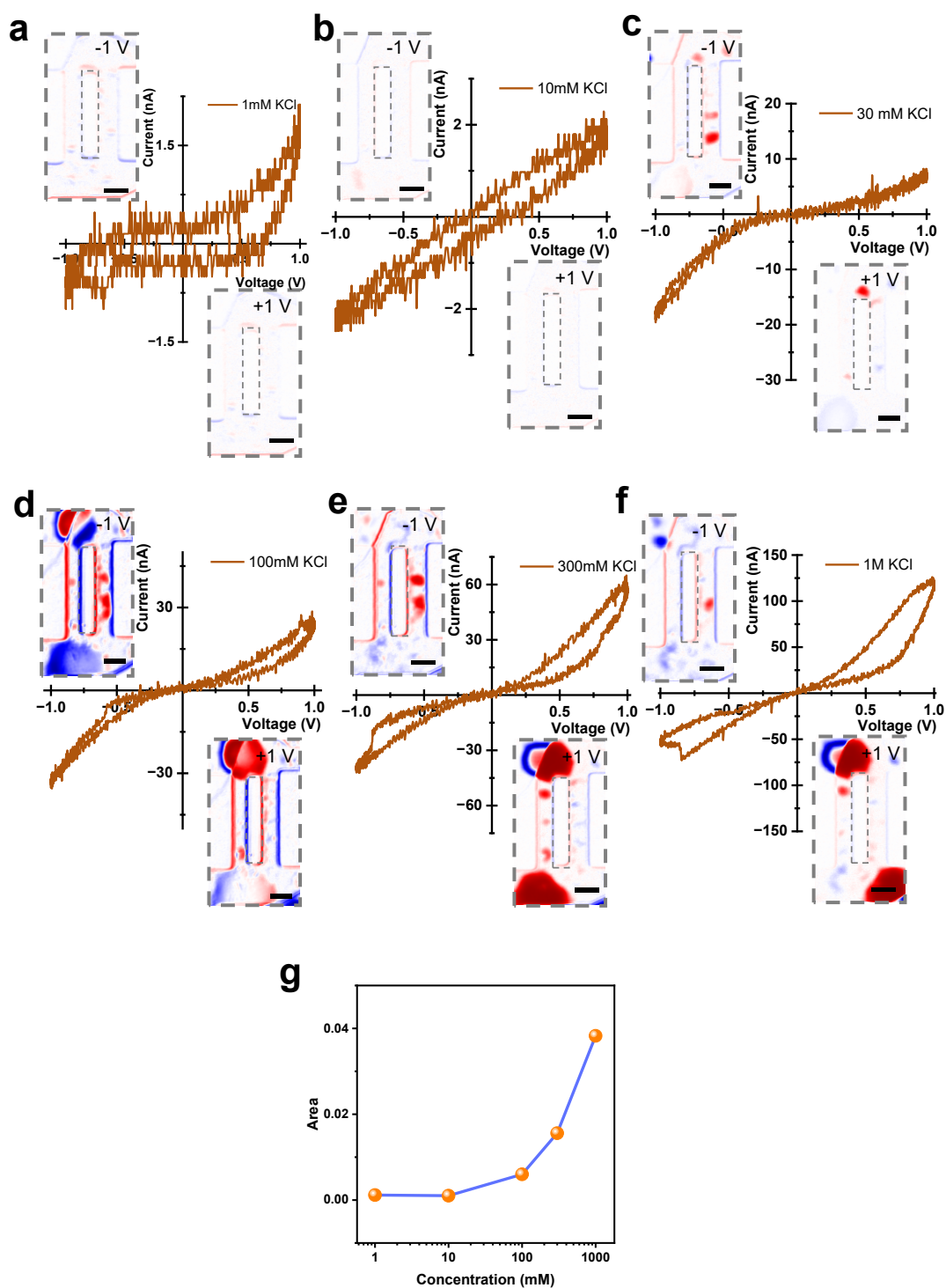
Supplementary Fig. 19 | I - V cycles of device 2 over 227 measurement cycles in 1 M KCl, recorded at ± 1 V and frequencies of 10 mHz and 100 mHz. Each panel shows a consecutive group of cycles, with earlier cycles in blue and later cycles in yellow. The dataset captures the full electrochemical history of the device providing a comprehensive picture of how memristive behaviour in nanochannel evolves under prolonged, repeated electrical stimulation. I - V traces are coloured blue to yellow in order of increasing cycle number.



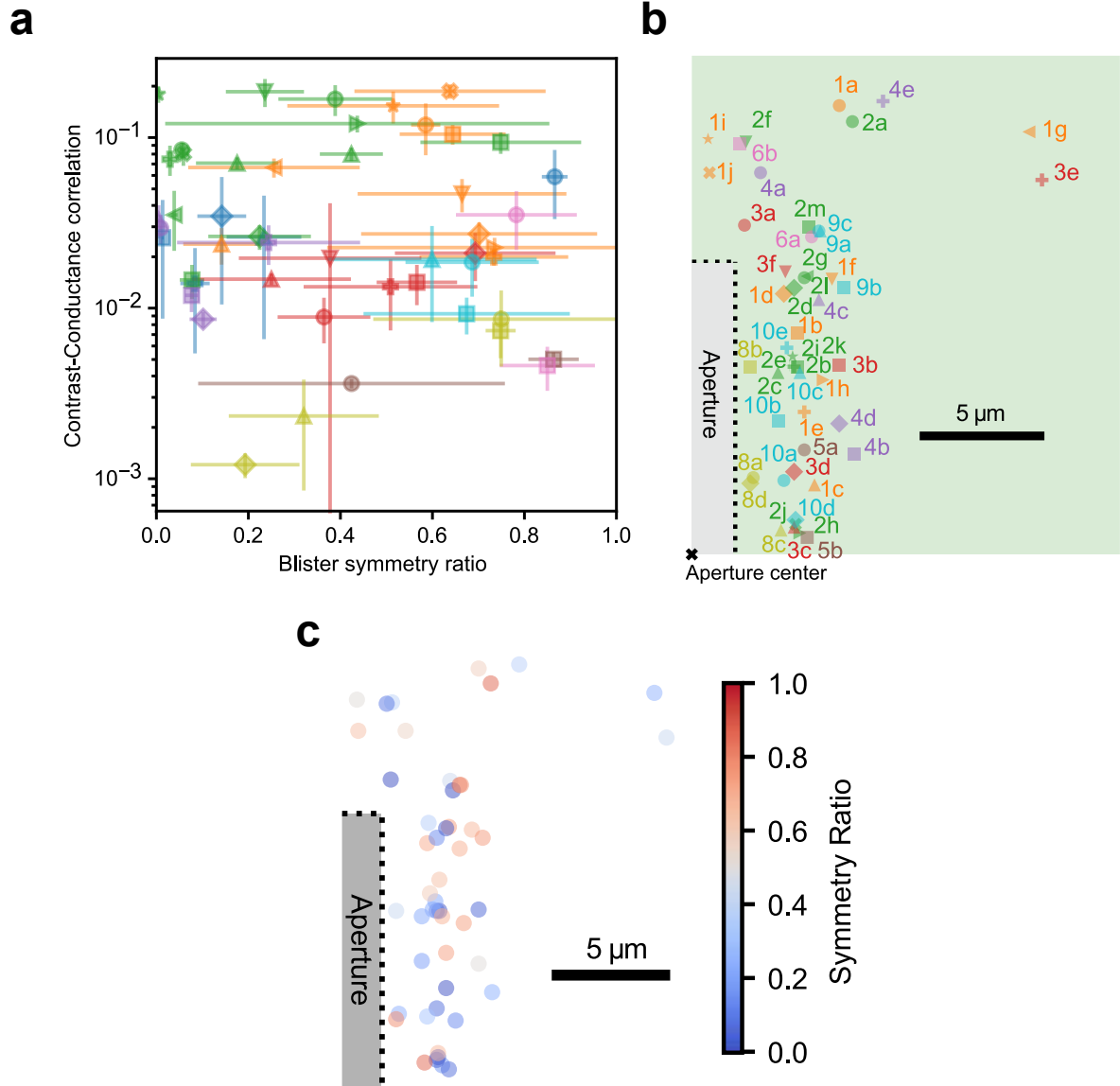
Supplementary Fig. 20 | Selected I-V cycles of Device 2 alongside VIC images at the corresponding cycle numbers. Cycles are drawn from the full 227 cycle data set in Supplementary Figure 19, recorded in 1 M KCl at ± 1 V and frequencies of 100 mHz (panels a, c-e) and 10 mHz (panel b and f). The selected cycles capture the transitions in I-V and the corresponding VIC images and show that these transitions coincide with the appearance of new blisters. The newly formed blisters are indicated by dashed brown circles in VIC images. In panel a, despite the voltage range of ± 1 V, the device exhibits a strictly linear off-state response (negative polarity) in the absence of any blister formation consistent with the low voltage response in Figure 2a confirming active structural evolution is required for hysteresis irrespective of the applied voltage. *I-V* traces are coloured blue to red in order of increasing cycle number. Scale bar: 5 μ m



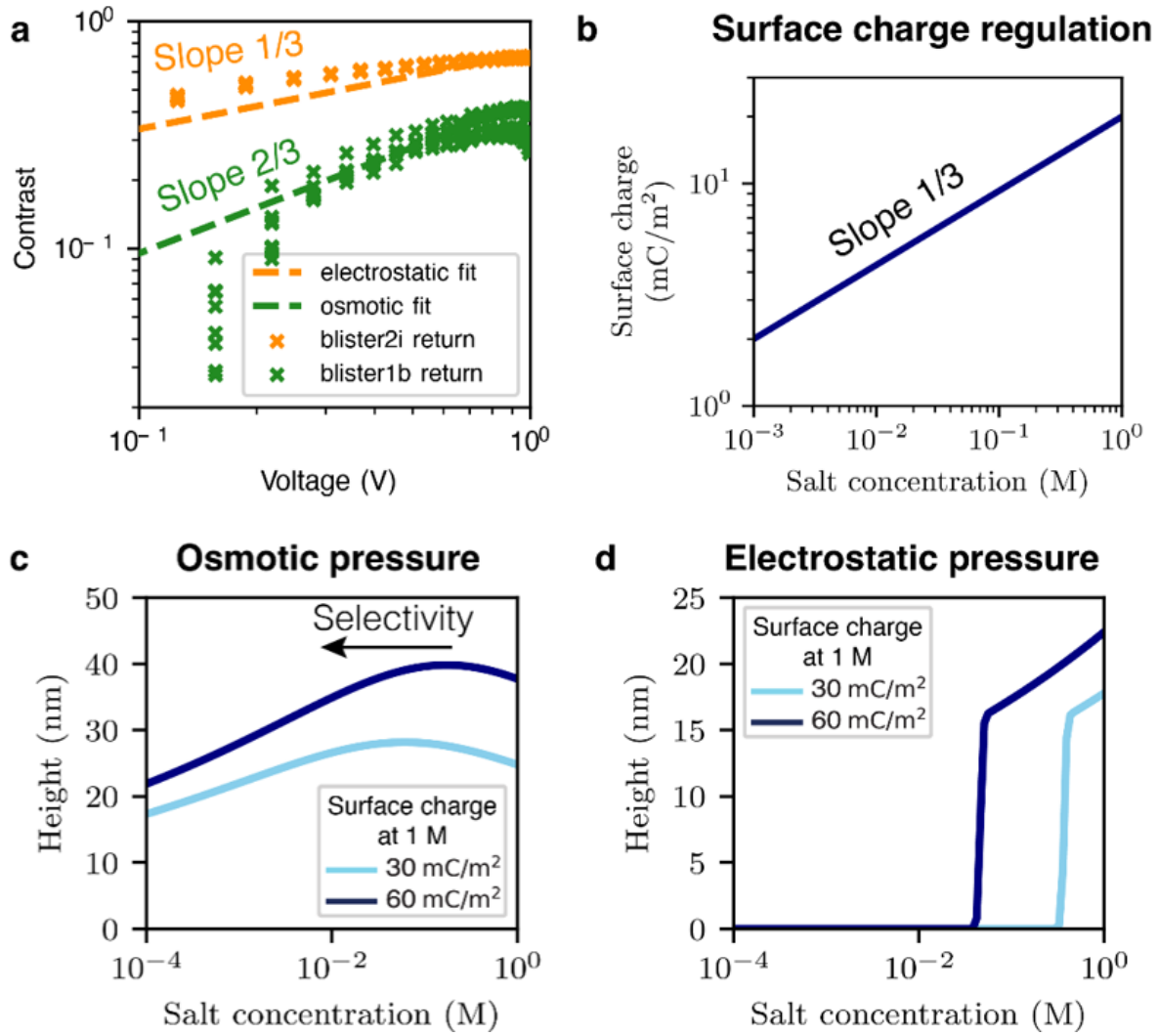
Supplementary Fig. 21 | Blisters can form at any interface of the heterostructure: I - V curves and VIC images and time traces of contrast of selected ROIs, device conductance and applied voltage for two devices measured in 1 M KCl at a frequency $f = 10$ mHz. (a) Device 9 (graphite bottom): the VIC images show blisters forming above the graphite layer, since the graphite is opaque and any deformation beneath it would not be optically visible. (b) Device 12 (hBN on SiN_x): blisters are visible on the bare hBN surface indicating that the delamination can also occur at the bottom interface. Together, panels a and b show that blister nucleation is not confined to the top surface and it can occur at any interface within the heterostructure (above or below). ROIs 1-3 (panel a) and ROIs 1-7 (panel b) mark the regions used to extract the optical contrast time traces. Scale bar: 5 μ m



Supplementary Fig. 22 | (a-f) I - V curves and VIC images of device 2 at ± 1 V with varying concentration of KCl from 1 mM to 1 M at a frequency $f = 10$ mHz and (g) shows corresponding hysteresis loop area vs concentration plot. At low concentrations (1-30 mM), the I - V is nearly linear with 30 mM showing rectification in I - V . and VIC images show little contrast. Both the hysteresis loop area and VIC increase above 100 mM with 1 M KCl producing the largest loop area. The concentration dependence in panel g indicates a threshold behaviour with memristive response emerging only once the ionic strength exceeds a critical value. Scale bar: 5 μ m



Supplementary Fig. 23 | (a) Scatter plot of C versus S for all blisters analyzed in hBN devices ($N = 50$ blisters from 9 devices) as shown in Fig. 5b with error bars (values obtained from $n = 2$ to $n = 4$ cycles), (b) Extended spatial distribution of blister positions relative to the aperture centre, overlaid for all devices and (c) Blister positions as (b) with each blister colour coded by its symmetry ratio S . Close to the aperture, both high and low symmetry blisters are found, whereas far from the aperture low-symmetry (unidirectional) blisters dominate. This observation is consistent with the osmotic model for bidirectional blisters, which requires the blister to be located on the ion transport path.



Supplementary Fig. 24 | **a**, Scaling of the contrast (or equivalently height) in the receding dynamics as a function of voltage for both models and for experimental data of model blisters (1b for the bidirectional memristor and 2i for the unidirectional memristor). **b**, Model used for the surface charge regulation: $\Sigma \sim c_o^{1/3}$. **c**, Prediction of maximum blister height as a function of salt concentration in the model of osmotic pressure. **d**, Prediction of maximum blister height as a function of salt concentration in the model of electrostatic pressure.

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